

**MICHIGAN MEMORIAL PHOENIX PROJECT  
PHOENIX MEMORIAL LABORATORY  
THE UNIVERSITY OF MICHIGAN**

**NEUTRON ABSORPTION IN PILE NEUTRON  
ACTIVATION ANALYSIS**

**OVE T. HOGDAHL**



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## ABSTRACT

As a basis for the discussion of the absorption effects in pile neutron activation analysis, this report begins with an expression for the pile neutron-induced activity in a nuclide by an  $(n, \gamma)$ -reaction under the assumption of no absorption effects. The induced activity is expressed in terms of a thermal neutron density and a pile neutron absorption cross-section or a resonance integral.

The absorption effects of pile neutrons on the neutron induced activity are discussed, and the activity is expressed in terms of an average thermal neutron density and an effective pile neutron absorption cross-section or an effective resonance integral.

The absorption effect of the thermal neutrons is measured by the ratio of the average thermal neutron density inside the sample to the unperturbed thermal neutron density at the site of the sample prior to its insertion into the neutron flux. A comparison between theoretical and experimental values for this ratio for spherical samples is given.

The effect of the absorption of the epithermal neutrons is shown to depend on the cadmium ratio and in a complex manner on the composition of the sample. A calculation of this effect on the pile neutron activation analysis of copper and gold in silver is given as an illustration of the calculations involved. The results are compared with experimental results.

Methods by which the effects of neutron absorption can be eliminated or experimentally determined are reviewed, and their limitations are discussed.



# NEUTRON ABSORPTION IN PILE NEUTRON ACTIVATION ANALYSIS

Ove T. Høgdahl\*

## 1. INTRODUCTION

The effects of the absorption of pile neutrons on the accuracy of a neutron activation analysis will depend on the samples and standards used, the preparation of these, and on the techniques by which the irradiations are carried out.

Fortunately, the effects of the neutron absorption are small in most neutron activation analysis, but it is necessary for the analyst to be able to calculate approximately the effects of neutron absorption in order to decide when these effects will be important. Though many practical methods exist by which the effects of the neutron absorption can be eliminated, the composition of the sample in some cases requires a careful consideration of the absorption effects in order to use these methods correctly.

The purpose of this report is to provide the reader with a basic understanding of the effects of neutron absorption in activation analysis, and to give relatively simple methods by which he will be able to approximately calculate these effects. The report does not intend to give a rigorous treatment of all aspects of the problem. Even with the most refined diffusion and transport theories which are used in reactor analysis, the conditions under

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which irradiation of a sample is generally carried out are so complex as to prohibit an accurate theoretical treatment. The sample is generally of a complex geometry, the immediate surroundings of the sample usually include various moderators, reflectors, coolant and fuel materials, and the site of the irradiation will often be in a channel in the reactor in which the neutron density varies spacially and in which the streaming of neutrons gives rise to quite different neutron currents incident on the various surfaces of the sample.

As a basis for the discussion of the absorption effects the report starts with a derivation of an expression for the pile neutron induced activity in a nuclide by an  $(n,\gamma)$ -reaction, under the assumption of no absorption effects. In the derivation it has been assumed that the absorption cross-section follows a pure  $\frac{1}{v}$ -law up to an energy of 0.4 eV, with resonance absorption peaks superimposed on the  $\frac{1}{v}$ -curve for energies above 0.4 eV. Further it has been assumed that a cadmium foil of a thickness of 0.01 in. will absorb all neutrons with energies below 0.4 eV and transmit all neutrons with energies above 0.4 eV.

The formula derived by Westcott, Walker and Alexander<sup>1</sup> takes into account both the deviation from the  $\frac{1}{v}$ -law in the thermal region and the actual energy dependence of the absorption cross-section for cadmium. Assuming the same total neutron density and the same energy distribution for the neutrons, the two formulas, in most cases, will give nearly the same result for the induced activity, and only in a few cases will the difference be as much as 10-15%.

The author recommends the method suggested by Westcott et al.,<sup>1</sup> for calculating induced activities in dilute samples (no absorption effects), but

has decided to use the simplified formula in the present discussion of the absorption effects because this simplifies the discussion to a great extent. As stated above no rigorous treatment of the absorption effects can be made. One must use simplified models and make assumptions which make the difference between the two formulas for induced activity insignificant.

In the derivation of the ratio of the average neutron density inside a sample to the neutron density at the site of the sample prior to its insertion into the reactor  $\frac{\bar{N}}{N}$ , we have assumed the sample to be a sphere with the same volume as the actual sample, and that it is in a non-scattering medium (poor moderator). The calculation of  $\frac{\bar{N}}{N}$  is based on the escape probabilities for spheres calculated by Case, deHoffmann and Placzek.<sup>2</sup> In cases where the shape of the sample deviates appreciably from a sphere, e.g., is in a form of a wire or a foil, one can expect that, depending on the geometrical shape, a better approximate value for  $\frac{\bar{N}}{N}$  can be found by treating the sample either as an infinite cylinder, infinite slab or a spheroid. Escape probabilities for these three geometries are given by Case et al.<sup>2</sup>

The assumption of a non-scattering medium around the samples is fairly good. Most often the samples are surrounded by air and/or aluminum in which the diffusion coefficient is so large as to justify the above assumption. If the sample is in contact with moderators like water or graphite, one must calculate  $\frac{\bar{N}}{N}$  by using methods which take the flux depression outside the sample into consideration. Meghreblian and Holmes<sup>3</sup> give formulas by which  $\frac{\bar{N}}{N}$  can be found for spheres, infinite slabs and infinite cylinders, but state that the formulas are best applied to absorbers with dimensions greater than

the diffusion length in the moderator. The diffusion lengths in water and graphite are 2.73 cm and 54.4 cm respectively.<sup>4</sup> Much theoretical and experimental work has been done to calculate the absorption effects in foils. The most important results of the theoretical calculations and experimental work can be found in the handbook "Reactor Physics Constants"<sup>4</sup> and in the papers by Sola,<sup>5</sup> Ritchie and Eldridge,<sup>6</sup> and Dalton and Osborne.<sup>7</sup>

The effective resonance integral is calculated by the method of Chernick and Vernon.<sup>8</sup> These calculations do not take the Doppler effect into consideration, and the resonances must be treated either as a "narrow resonance (NR)" or a "narrow infinitely heavy absorber (NRIA)." The reason for using this method is that Chernick and Vernon<sup>8</sup> give reasonably simple analytical expressions by which approximate values for the effective resonance integral can be calculated. In most cases one must make several assumptions prior to the calculation of the effective resonance integral, so in most cases, more accurate and sophisticated methods are not justified.

## 2. DISINTEGRATION RATE DUE TO THE $(n,\gamma)$ -REACTION

### 2.1 THE NEUTRON SPECTRUM

We will assume that the neutron spectrum is composed of two parts, a Maxwellian distribution corresponding to  $T^\circ\text{K}$ , cut off at a suitable higher limit of velocity equal to  $v_2$ , and an epithermal  $\frac{dE}{E}$ -distribution cut off at a lower limit of velocity corresponding to the same velocity  $v_2$ . We will set  $v_2$  equal to  $v_{T\mu}^{1/2}$ , where  $v_T$  is equal to  $v_0(T/T_0)^{1/2}$ .  $v_0$  (2200 m/sec) is the velocity of a neutron of energy  $kT_0$ , where  $T_0 = 293.6^\circ\text{K}$ . For the reactors

"JEEPI," which are situated near Oslo, Norway, "RI" in Stockholm, Sweden, and "BEPO," Harwell, England, it can be found by using the data given by Grimeland,<sup>9</sup> Johansson, Lampa and Sjöstrand<sup>10</sup> and Taylor,<sup>11</sup> that the value for  $\mu$  is about 5. "JEEPI" and "RI" are heavy water moderated reactors, while "BEPO" is graphite moderated.

The assumed neutron density distribution per unit velocity interval may be written:

$$n(v) = n_m \rho_m(v) + n_e \rho_e(v) \quad (1)$$

where  $\rho_m(v)$  and  $\rho_e(v)$  are the Maxwellian and epithermal density distribution functions normalized so that

$$\int_0^{\infty} \rho_m(v) dv = \int_0^{\infty} \rho_e(v) dv = 1$$

$n_m$  is the density of the neutrons in the Maxwellian part of the spectrum and  $n_e$  is the density of the epithermal neutrons. By the normalizing conditions, the total neutron density,  $n$ , equal to the sum of  $n_m$  and  $n_e$ , can be written:

$$n = n_m + n_e = \int_0^{\infty} n(v) dv \quad (1a)$$

The normalized form of  $\rho_m(v)$  and  $\rho_e(v)$  are:

$$\rho_m(v) = \frac{C}{v_T^3} \cdot v^2 \cdot e^{-\left(\frac{v}{v_T}\right)^2}$$

where  $C = 2.6$  for  $\mu = 5$

$$\rho_e(v) = v_T^{\mu-1/2} \cdot \frac{1}{v^2}$$

The reason for ignoring the presence of a fission spectrum is that the contribution to the total  $(n,\gamma)$ -reaction rate due to fission neutrons usually is very small.

## 2.2 ABSORPTION CROSS-SECTION FOR PILE NEUTRONS

By irradiating  $N_A$  nuclides of the type A in the neutron spectrum given by Eq. (1), the reaction rate per nuclide is given by:

$$-\frac{1}{N_A} \frac{dN_A}{dt} = \int_0^{\infty} n(v) v \sigma(v) dv \quad (2)$$

where  $\sigma(v)$  is the absorption cross-section for the reaction  $A(n,\gamma)B$ . For nearly all the nuclides used in pile neutron activation analysis,  $\sigma(v)$  can be written:

$$\sigma(v) = \frac{\sigma_0 v_0}{v} + \sigma_r(v) \quad (3)$$

where  $\sigma_0$  is the absorption cross-section for neutrons with a velocity of 2200 m/sec, and where  $\sigma_r(v)$ , which usually is zero for neutron energies below 0.4 eV, takes into account resonance absorptions.

Equation (2) can be written:

$$-\frac{1}{N_A} \cdot \frac{dN_A}{dt} = \int_0^{v_1} n(v) v \sigma(v) dv + \int_{v_1}^{\infty} n(v) v \sigma(v) dv \quad (4)$$

where  $v_1$  corresponds to an energy of 0.4 eV ( $8.75 \cdot 10^3$  m/sec). For nuclides which follow the  $\frac{1}{v}$ -law fairly closely for energies below 0.4 eV the first integral in Eq. (4) is equal to:

$$\sigma_0 v_0 \int_0^{v_1} n(v) dv = \sigma_0 v_0 N_{th} \quad (5)$$

where  $N_{th}$  is the density of neutrons with energies up to 0.4 eV.  $N_{th}$  will be called the thermal neutron density. Note that the contribution to the total reaction rate due to the thermal neutrons is independent of the neutron energy distribution, only proportional to the thermal neutron density.

Because  $v_1$  usually is larger than the cut off velocity  $v_2$  defined in Section 2.1, the second integral in Eq. (4) can be written:

$$n_e v_T \mu^{1/2} \int_{v_1}^{\infty} \frac{\sigma(v)}{v} dv = \frac{n_e v_T \mu^{1/2}}{2} \int_{0.4 \text{ eV}}^{\infty} \frac{\sigma(E)}{E} dE. \quad (6)$$

The integral above is the sum of two components, one corresponding to the  $\frac{1}{v}$  part of the cross-section and one part corresponding to the resonance absorption. The first part is equal to  $0.5 \sigma_0$ . When a cadmium foil (0.01 in.-0.02 in. thick) is used to filter off the thermal neutrons, the value is less than  $0.5 \sigma_0$ . The actual value is of little importance. For a  $\frac{1}{v}$ -nuclei we have used the same value,  $0.44 \sigma_0$ , as used by Macklin and Pomerance.<sup>12</sup> The second part is the resonance integral:

$$J = \int_{0.4 \text{ eV}}^{\infty} \frac{\sigma_r(E)}{E} dE.$$

With the help of Eqs. (5) and (6), Eq. (4) can be written:

$$-\frac{1}{N_A} \cdot \frac{dN_A}{dt} = \sigma_0 v_0 N_{th} + \frac{n_e v_T \mu^{1/2}}{2} (J + 0.44 \sigma_0)$$

or

$$-\frac{1}{N_A} \cdot \frac{dN_A}{dt} = \sigma_{pile}^A \cdot v_0 \cdot N_{th} \quad (7)$$

where

$$\sigma_{pile}^A = \sigma_0 + \frac{n_e v_T \mu^{1/2}}{2 v_0 N_{th}} (J + 0.44 \sigma_0). \quad (8)$$

### 2.3 THE DISINTEGRATION RATE

The number of nuclides B formed per unit time by the reaction  $A(n, \gamma)B$  can be written:

$$\frac{dN_B}{dt} = N_A \sigma_{\text{pile}}^A v_0 N_{\text{th}} - N_B (\lambda_B + \sigma_{\text{pile}}^B v_0 N_{\text{th}}) \quad (9)$$

where  $\sigma_{\text{pile}}^B$  is the pile absorption cross-section and  $\lambda_B$  the disintegration constant for the nuclide B, and where  $N_B$  is the number of nuclides B.

Because  $e^{-\sigma_{\text{pile}}^A v_0 N_{\text{th}}}$  is approximately unity, and the products  $\sigma_{\text{pile}}^A v_0 N_{\text{th}}$  and  $\sigma_{\text{pile}}^B v_0 N_{\text{th}}$  usually are small compared to  $\lambda_B$ , one gets with a good approximation the following solution of the differential Eqs. (7) and (9):

$$\lambda_B N_B = N_A^0 \sigma_{\text{pile}}^A v_0 \cdot N_{\text{th}} (1 - e^{-\lambda_B t})$$

where  $N_A^0$  is the number of the nuclides A before the irradiation.

We will write the equation above in terms of the disintegration rate of the nuclides B at the end of the irradiation,  $D_B^0$ , and will express  $N_A^0$  in terms of the weight of the irradiated nuclides A:

$$D_B^0 = \frac{x}{M} \rho \mathcal{N} \sigma_{\text{pile}}^A v_0 N_{\text{th}} (1 - e^{-\lambda_B t}) \quad (10)$$

where

$x$  = grams of the element, with atomic weight  $M$ , which contains  $N_A^0$  nuclides of the type A.

$\rho$  = isotopic abundance

$\mathcal{N}$  = Avogadro's number

$t$  = the length of irradiation.

## 2.4 CADMIUM RATIO

The cadmium ratio of the nuclei A is defined as the ratio of the induced specific activity of the nuclides B in a bare sample to the induced specific activity of B in a cadmium covered sample. No absorption effects may be present.

$$R = \frac{\sigma_0 v_0 N_{th} + \frac{n_e v_{T\mu}^{1/2}}{2} (J + 0.44\sigma_0)}{\frac{n_e v_{T\mu}^{1/2}}{2} (J + 0.44\sigma_0)}$$

or

$$R - 1 = \frac{\sigma_0 v_0 N_{th}}{\frac{n_e v_{T\mu}^{1/2}}{2} (J + 0.44\sigma_0)} \quad (11)$$

Combining Eqs. (8) and (11):

$$\sigma_{pile}^A = \sigma_0 \frac{R}{R-1} \quad (12)$$

## 3. THE EFFECTS OF NEUTRON ABSORPTION

### 3.1 THE NEUTRON SPECTRUM

The neutron density will generally decrease inside a sample placed in a neutron bath (self-shielding). Because the absorption cross-section is a function of the velocity, the shape of the spectrum will change within the sample.

If we look at the Maxwellian part of the spectrum, the mean velocity will increase with increasing distance into the sample. We get a "hardening of the flux,"<sup>13</sup> caused by the fact that the slowest neutrons will be absorbed more strongly than neutrons with higher velocities ( $\frac{1}{v}$ -law for the absorption cross-section).

The same change in the spectrum towards higher energies will also take place in the epithermal part of the spectrum, but if the bulk part of the sample contains elements which have nuclides with high resonance absorptions, the greatest change in the epithermal part of the spectrum will be caused by absorption of resonance neutrons. An indium foil with a thickness of 0.02 gm indium/cm<sup>2</sup>, placed in a beam of epithermal neutrons, will for example nearly completely remove neutrons with energies near 1.46 eV.<sup>13</sup> This energy corresponds to the largest resonance absorption in In<sup>115</sup>.

The neutron density distribution per unit velocity interval inside a sample, is therefore not only a function of the neutron velocity but also a function of the space coordinates. It should be obvious from the above discussion that it is very difficult to write down the general analytical expression for the neutron density distribution function inside a given sample.

### 3.2 EFFECTIVE ABSORPTION CROSS-SECTION FOR PILE NEUTRONS

From Eq. (4) we can see that the reaction rate  $\frac{dN_A}{dt}$  is the sum of two terms, one term associated with the thermal part of the spectrum (neutrons with energies up to 0.4 eV), and one term associated with the epithermal part:

$$\frac{dN_A}{dt} = \left(\frac{dN_A}{dt}\right)_{th} + \left(\frac{dN_A}{dt}\right)_e \quad (13)$$

Because we have assumed that the absorption cross-section follows a  $\frac{1}{v}$ -law up to an energy of 0.4 eV,  $\left(\frac{dN_A}{dt}\right)_{th}$  will be independent of the velocity distribution of the neutrons. The term is only proportional to the density of the thermal neutrons, and one can easily show that:

$$-\frac{1}{N_A} \left( \frac{dN_A}{dt} \right)_{th} = \sigma_0 v_0 \bar{N}_{th} \quad (14)$$

where  $\bar{N}_{th}$  is the average thermal neutron density inside the sample.

It is extremely difficult to write down a general expression for  $\left( \frac{dN_A}{dt} \right)_e$  and we will only define an effective resonance integral,  $J^{eff}$ , by the equation:

$$-\frac{1}{N_A} \left( \frac{dN_A}{dt} \right)_e = \frac{n_e v_{T\mu}^{1/2}}{2} (J^{eff} + 0.44\sigma_0) \quad (15)$$

Using Eqs. (13), (14) and (15) we can now define an effective absorption cross-section for pile neutrons by the equation:

$$-\frac{1}{N_A} \frac{dN_A}{dt} = \sigma_{pile}^{eff} \cdot v_0 \bar{N}_{th} \quad (16)$$

where

$$\sigma_{pile}^{eff} = \sigma_0 + \frac{n_e v_{T\mu}^{1/2}}{2 v_0 \bar{N}_{th}} (J^{eff} + 0.44\sigma_0) \quad (17)$$

### 3.3 THE DISINTEGRATION RATE

As in Section 2.3 we can write the disintegration rate of the nuclides B at the end of the irradiation as:

$$D_B^0 = \frac{x}{M} \cdot \rho \cdot \mathcal{V} \cdot \sigma_{pile}^{eff} v_0 \bar{N}_{th} (1 - e^{-\lambda_B t}) \quad (18)$$

### 3.4 THE AVERAGE THERMAL NEUTRON DENSITY

Much confusion exists concerning self absorption, flux depression and flux perturbation, and it will be useful to point out the differences between these terms. When an absorber is placed in a diffusion media, the neutron flux inside and outside the sample will be perturbed. In the vicinity of

the absorber there will exist a flux gradient such that the neutron flux decreases in the direction towards the sample. This flux depression effect is caused by the fact that neutrons which are absorbed in the sample will be unable to scatter back into the moderator and maintaining a constant neutron flux. The neutron flux will decrease inside the sample because neutrons are removed from the flux by absorption. This effect is called the self absorption effect while the total effect of the flux depression and self absorption is called the flux perturbation effect. If the neutron flux of the unperturbed flux is called  $\phi$ , the average flux over the surface of the sample for  $\phi_s$ , and the average flux inside the sample for  $\bar{\phi}$ , one can define these three factors:

$$\begin{aligned} \text{Flux depression factor: } & \frac{\phi_s}{\phi} \\ \text{Self absorption factor: } & \frac{\bar{\phi}}{\phi_s} \\ \text{Flux perturbation factor: } & \frac{\bar{\phi}}{\phi} \end{aligned}$$

As mentioned in the introduction, the conditions under which an irradiation of a sample generally is carried out are so complex that an accurate theoretical calculation of the three absorption factors defined above is prohibitive. As a measure of the absorption effect on the neutron density we will in this report use the ratio of the average thermal neutron density inside a sample to the thermal neutron density at the site of the sample prior to its insertion into the reactor  $\left(\frac{\bar{N}_{th}}{N_{th}}\right)$ . By thermal neutrons we will, as before, understand neutrons with an energy below 0.4 eV. To be able to get approximate values for this ratio we will choose the following simple model. The samples which are assumed to be of spherical geometry with the same volume

as the actual sample are immersed in a homogeneous non-scattering medium where we have an isotropic neutron flux. The calculations will be based on the work of Case et al.<sup>2</sup> It should be noted that the thermal neutron density,  $N_{th}$ , for a well moderated reactor is responsible for more than 90% of the total neutron density [as defined by Eq. (1a)]. From Eq. (11) one can find that even in an irradiation position where the cadmium ratio for gold is as low as 2.5, the ratio of  $n_e$  to  $N_{th}$  will be about 0.03. It should also be pointed out that when the samples are surrounded by materials which have a large diffusion coefficient, the effect of the flux depression will be small. However, when the sample is surrounded by a good moderator as water or graphite, this effect can be considerable. Levin and Hughes<sup>14</sup> found that the flux in the vicinity of a cylindrical cobalt shell 12-1/4 in. long, with an outside diameter of 1.045 in. and a 0.125 in. wall thickness, surrounded by graphite, was perturbed up to a distance of about 14 in. Høgdahl<sup>15</sup> found, however, that when a silver sphere with a diameter of 6.4 mm was placed in an aluminum box about 3.2 in. long and with an outside diameter of about 1 in., the flux was depressed only at distances less than 7-10 mm. In the experiment, the box was filled with aluminum foil and irradiated in a channel in the reactor "JEEPPI," Norway, at a point where the flux gradient was found to be 0.8% per cm along the center line of the channel.

If a pure absorbing material is located in an isotropic neutron bath (non-scattering medium) of monoenergetic neutrons, Case et al.,<sup>2</sup> have shown that the ratio of the average neutron density inside the absorber to the neutron density at a large distance outside the material is  $P_0$ , the escape

probability. Values of  $1-P_0$ , the collision probability, have been tabulated<sup>2</sup> for spherical objects as a function of the ratio of the objects radius to the absorption mean-free-path in the absorber.

An actual sample is not a pure absorber, but Case et al.,<sup>2</sup> have shown that the scattering cross-section must be larger than the absorption cross-section before the scattering will have any pronounced effect on  $P_0$ .

Case et al.,<sup>2</sup> also give an expression for  $P_0$  for oblate spheroids and discuss the dependence of escape probability on shape. They have found that the major axis,  $a$ , must be much larger than the minor axis,  $b$ , before the escape probability will differ appreciably from that of a sphere of the same volume. For two oblate spheroids, for example, with  $\frac{a}{b}$  equal to  $\frac{5}{2}$  and 5 respectively, and where  $\frac{\bar{N}}{N}$  is larger than 0.7 (less than 30% absorption of neutrons), the difference between  $P_0$  and the escape probability of a sphere of the same volume will be less than 3% and 7% respectively.

As mentioned earlier the values for  $P_0$  for spherical samples are calculated as a function of the ratio of the radius of the spheres to the absorption mean-free-path.<sup>2</sup> They assumed monoenergetic neutrons, but the value for  $\frac{\bar{N}_{th}}{N_{th}} (P_0)$  can be found by calculating the absorption mean-free-path by the equation:

$$1 = \frac{1}{\sum n_i \bar{\sigma}_i} \quad (19)$$

where  $n_i$  = number of nuclides of type  $i/\text{cm}^3$

$\bar{\sigma}_i$  = the average absorption cross-section for nuclides of the type  $i$ .

$\bar{\sigma}_i$  is defined by the equation:

$$\bar{\sigma}_i = \frac{\int_0^{v_1} n(v) v \sigma_i(v) dv}{\int_0^{v_1} n(v) v dv} \quad (20)$$

where  $v_1$ , as before, corresponds to a neutron energy of 0.4 eV.

If the analytical expression for  $n(v)$  is unknown, an approximate value for  $\bar{\sigma}_i$  can be obtained by assuming  $n(v)$  to be a Maxwellian distribution function and setting  $v_1$  equal to infinity. In this case

$$\bar{\sigma}_i = \sigma_{0i} \frac{\sqrt{\pi}}{2} \cdot \sqrt{\frac{293.6}{T}} \quad (21)$$

where  $\sigma_{0i}$  is the absorption cross-section for neutrons with a velocity of 2200 m/sec.

For a well-moderated reactor, the difference between Eqs. (20) and (21) will be small. For the reactor "JEEPI," Norway, which is a heavy water moderated uranium reactor (natural uranium), the difference is only 1.2%.

As mentioned in Section 3.1 an absorber will cause a "hardening of the flux" because the slowest neutrons will be absorbed more strongly than neutrons with higher velocities. The effect of this hardening of the flux on the effective cross-section will depend on  $\sigma_i(v)$ . For a  $\frac{1}{v}$ -cross-section the actual effective cross-section will be smaller than the value calculated by Eq. (21). Bethe<sup>16</sup> has calculated the hardening effect for nuclides which follow the  $\frac{1}{v}$ -law. He assumes a beam of neutrons incident normally on an absorbing slab. From a graph<sup>16</sup> one can read that with a transmission of 0.7, the measured cross-section will be about 3% less than the value one would have measured if there were no hardening effect. Westcott<sup>17</sup> has calculated

the hardening effect for several nuclides which do not follow the simple  $\frac{1}{v}$ -law. He has assumed either a beam of neutrons incident normally on an absorbing slab, or isotropic incidence on one side of the slab. From the graphs<sup>17</sup> one has read that for both samarium and cadmium, which deviate appreciably from the  $\frac{1}{v}$ -law, the effective cross-section for the hardened spectrum,  $\hat{\sigma}_H$ , at a transmission of 0.7 is about 6% larger than the effective cross-section for the unhardened spectrum,  $\hat{\sigma}_M$ . For gadolinium, which also deviated from the  $\frac{1}{v}$ -law, the  $\hat{\sigma}_H$ , at a transmission of 0.7 is about 1% smaller than  $\hat{\sigma}_M$ . These values are for a neutron temperature of 20°C and for an isotropic incidence of neutrons on one side of a slab.

Although it is not straightforward to compare beam geometry, or even isotropic incidence on one side of a slab, with irradiations of comparable small absorbing spheres in the reactor, the referred values indicate the amount of error introduced in  $\bar{\sigma}_i$  by using Eq. (21).

Høgdahl<sup>15</sup> has determined  $\frac{\bar{N}_{th}}{N_{th}}$  for spherical samples of cobalt, gold, silver and tungsten, and compared the experimental values with the theoretical values obtained by using Eqs. (19) and (21) and the tabulated values for  $P_0$  given by Case et al.<sup>2</sup> The reactors "JEEPI," Kjeller, Norway, "RI," Stockholm, Sweden, and "BEPO," Harwell, England, were used in the experiments. Both pure thermal and pile neutrons were used. The results are shown in Figure 1.

Very few experiments have been done to compare experimental values of  $\frac{\bar{N}_{th}}{N_{th}}$  for non-spherical samples with calculated values found by treating the samples as spheres with the same volume as the actual samples. The method has only been checked for five samples of powdered cobalt. The samples were

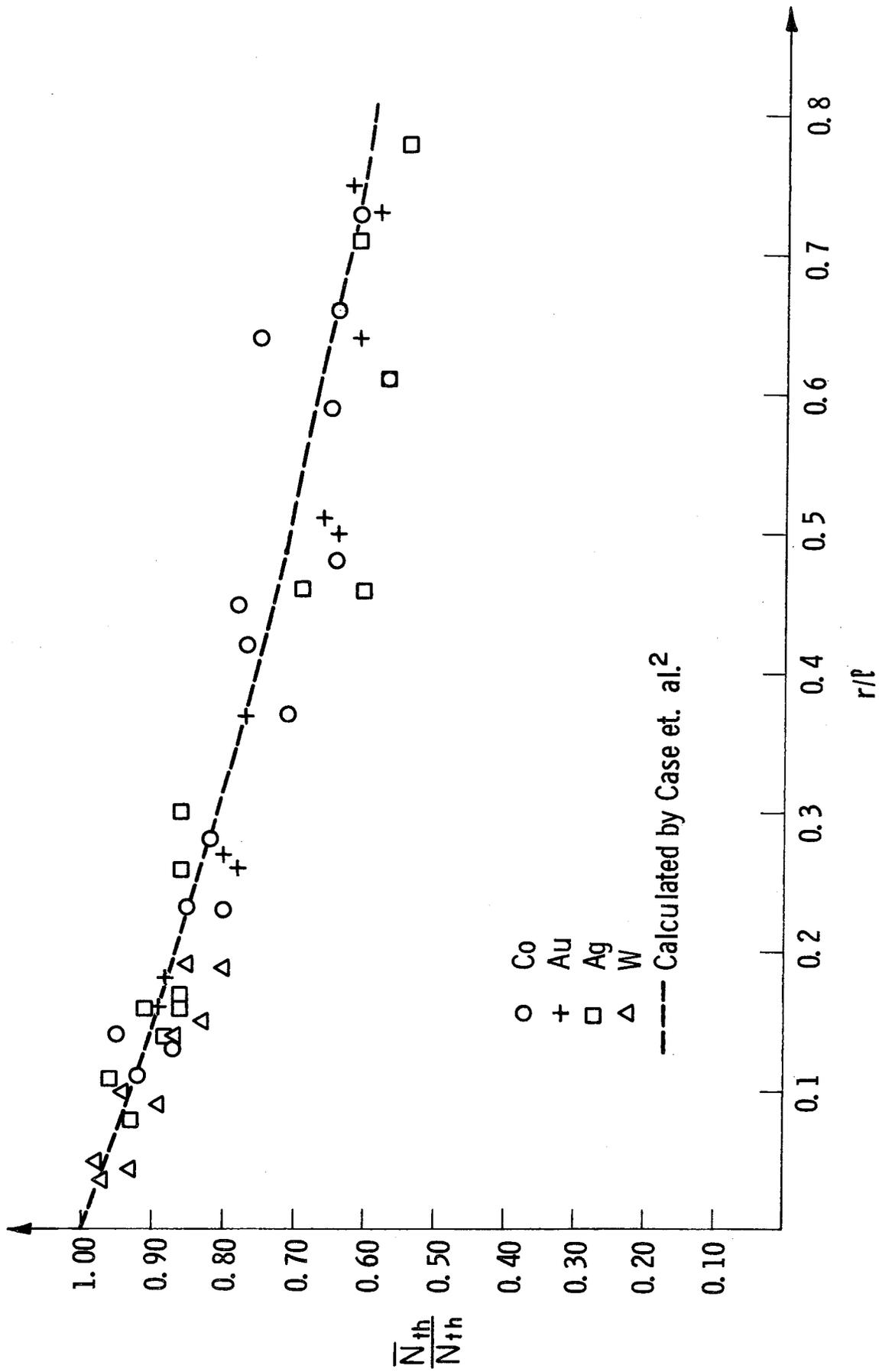


Figure 1.  $\bar{N}^{\text{th}} / N^{\text{th}}$  for spherical samples as a function of the ratio of the radius to the absorption mean-free-path.<sup>15</sup>

weighed into polyethylene tubes with an internal diameter of 5.3 mm, the powder was pressed together by means of a piece of cork, and the tubes were sealed. The specific weight of the powder in the samples was found to be 2.1 gm/cm<sup>3</sup> by calculating the volume occupied by one sample weighing 919 mg by weighing a similar tube filled with water to the same height as the cobalt sample. A cobalt standard solution containing 2 mg cobalt/ml and the five samples were irradiated with gold monitoring foils for 10 sec in the pneumatic tube system of the Ford Nuclear Reactor of The University of Michigan. The samples were allowed to "cool" for two days before the Co<sup>60</sup>-activity was measured with a gamma-ray spectrometer. Because the cadmium ratio of cobalt at the irradiation position was about 50, the ratio of the specific cobalt activity of the samples to the specific activity of the standard can be set equal to the ratio  $\frac{\bar{N}_{th}}{N_{th}}$ . The results of the experiments are shown in Table I.

Table I. Neutron Absorption of Powdered Cobalt Samples.

Weight (mg)	"Radius" (mm)	$\frac{\bar{N}_{th}}{N_{th}}$ (Calculated)	$\frac{\bar{N}_{th}}{N_{th}}$ (Experimental)
9.8	1.04	0.95	0.99
23.1	1.38	0.94	0.96
63.4	1.93	0.92	0.91
222.7	2.94	0.88	0.84
491.6	3.83	0.84	0.76

In cases where the shape of the sample deviates appreciably from a sphere, e.g., is in the form of a wire or a foil, one can expect that, depending on the geometrical shape, a better approximate value for  $\frac{\bar{N}_{th}}{N_{th}}$  can be found by treating the sample either as an infinite cylinder, infinite slab or a spheroid. Escape probabilities for these three geometries are also given by Case et al.<sup>2</sup>

### 3.5 THE EFFECTIVE RESONANCE INTEGRAL

The theory of resonance absorption was developed during the days of the Manhattan Project by Wigner.<sup>18</sup> His work anticipated most of the later developments. The first reasonably successful attempt to calculate effective resonance integrals for homogeneous assemblies by using resonance data was made in 1956 by Dresner.<sup>19</sup> This work was followed in 1958 by three independent calculations of resonance integrals for heterogeneous assemblies by Adler, Hinman, and Nordheim,<sup>20</sup> Dresner,<sup>21</sup> and by Chernick and Vernon.<sup>8</sup> These calculations give very similar results, but they differ in the mathematical approximations. Chernick and Vernon,<sup>8</sup> and to a lesser extent Dresner,<sup>21</sup> try to use analytical procedures as much as possible, whereas Adler, Hinman, and Nordheim<sup>20</sup> give a set of tables, from which the effective resonance integrals can be interpolated for a wide variety of geometries and temperatures. For all three calculations the wide low-lying resonances (low energies) are treated in the "narrow infinitely heavy absorber (NRHA)" approximation and the narrow resonances in the "narrow resonance (NR)" approximation. The distinction between a wide and a narrow resonance is generally made according to whether the practical width is smaller or larger than the maximum energy loss in a

collision. The practical width is the energy interval in which the absorption cross-section is larger than the potential scatter cross-section.

An improved numerical method has been invented by Nordheim.<sup>22</sup> By the use of a computer, FORTRAN IBM-7090, the effective resonance integrals can be calculated from information concerning the resonance parameters and specifications for temperature, composition, and geometry. Since the method is a numerical one, it has been possible to include effects which are otherwise difficult to treat, e.g., the Doppler effect and interference between resonance absorption and potential scattering.

An alternative method, which makes it possible to treat cases of arbitrary complication, is the Monte Carlo method. It has been successfully applied to resonance absorption by Richtmeyer.<sup>23</sup>

The methods mentioned above have been used to calculate the effective resonance integrals for nuclides which are important in reactor technology. The classical example is the calculation of the effective resonance integral for  $U^{238}$  in pure uranium metal, uranium oxide and uranium mixed with a moderator. This effective resonance integral is less than the dilute resonance integral (no absorption effect) primarily because of a "self-absorption" effect. Neutrons with energies within the resonance regions are absorbed primarily by the  $U^{238}$ -nuclides itself. In an activation analysis the picture is usually more complicated. The effective resonance integral for the nuclide of interest is often less than the dilute integral because of neutron absorption in the nuclides which make up the main constituents of the sample. The differences between the effective and dilute resonance integral are usually

small, but in cases where the bulk materials have absorption resonance peaks which coincide with resonance peaks in the nuclide of interest, the effect of the absorption of epithermal neutrons can be quite large. As an illustration we will consider the effective resonance integral for gold in a sample of silver and cobalt. The energy for the main resonance peak in gold which is responsible for about 92% of the total resonance integral, is 4.91 eV, while the energy for the main resonance peak in silver is 5.10 eV (resonance in  $\text{Ag}^{109}$ ). Even if the gold content is of the order of a few ppm, the effective resonance integral for gold will decrease rapidly with the sample size. The neutrons with energies around 4.91 eV are rapidly removed from the flux because of resonance absorption in  $\text{Ag}^{109}$ . For example, the effective resonance integral for gold in a silver sphere weighing only 48 mg, is 90% lower than the dilute integral<sup>15</sup> (see also Section 3.7 in this report). On the other hand, if we have microgram amounts of gold in cobalt, which has its main resonance peak at 132 eV, the neutrons with energies around 4.91 eV will be slowly removed from the flux by a  $\frac{1}{V}$ -absorption in the cobalt nuclides. By using the method outlined in Section 3.4, we find that only about 3% of the neutrons with energies around 4.91 eV (4.7-5.1 eV) are absorbed in a cobalt sphere weighing 1000 mg. In this case, therefore, the effective resonance integral for gold at 4.91 eV is only a few percent lower than the dilute integral.

### 3.6 THE EFFECTS OF EPITHERMAL NEUTRON ABSORPTION

The effective resonance integral  $J^{\text{eff}}$ , defined in Eq. (15), takes both resonance and  $\frac{1}{V}$ -absorption effects into account. In the equation mentioned,

the contribution to the induced activity from  $\frac{1}{V}$ -absorption of epithermal neutrons,  $\frac{n_e v_T \mu^{1/2}}{2} 0.44 \sigma_0$ , is the same as if no absorption effects are present.

We use the unperturbated epithermal neutron density,  $n_e$ , and all epithermal absorption effects must therefore be accounted for by  $J^{\text{eff}}$ . We have written Eq. (15) in the present form to get the expression for  $\sigma_{\text{pile}}^{\text{eff}}$  in Eq. (17) to be of the same form as the expression for  $\sigma_{\text{pile}}$  in Eq. (8). An alternative way to write Eq. (15) is

$$-\frac{1}{N_A} \left( \frac{dN_A}{dt} \right)_e = \int_V \int_{v_1}^{\infty} n(V, v) v \sigma(v) dv dV \quad (22)$$

where we must integrate over the energy range from 0.4 eV to infinity and over the whole volume of the sample. The function  $n(V, v)$  will depend markedly on the constituents of the sample, and will, for example, distort the simple  $\frac{dE}{E}$ -epithermal spectrum to a great extent if the nuclides which make up the bulk materials in the sample have several large resonance peaks.

The complexity of the problem of calculating the effects of absorption of epithermal neutrons abandons an accurate theoretical treatment. In some cases we can calculate the absorption effects of epithermal neutrons within 5-10%, but in many cases only a rough estimate of these effects can be obtained.

In an activation analysis we are measuring the total induced activity, that is, the sum of the activity induced by thermal and epithermal neutrons respectively. The effect of the absorption of epithermal neutrons on the total absorption effect will therefore depend on the degree of thermalization of the neutrons; that is, depend on the cadmium ratio at the irradiation

position. It is very useful to consider the ratio of the activity induced by epithermal neutrons to the activity induced by thermal neutrons. From Eqs. (11), (14) and (15) we get

$$\frac{\left(\frac{dN_A}{dt}\right)_e}{\left(\frac{dN_A}{dt}\right)_{th}} = \frac{(J^{eff} + 0.44\sigma_0)}{(J + 0.44\sigma_0)(R-1) \frac{\bar{N}_{th}}{N_{th}}} \quad (23)$$

In practice we have

$$0.5 < \frac{\bar{N}_{th}}{N_{th}} \leq 1 \quad \text{and} \quad (J^{eff} + 0.44\sigma_0) \leq (J + 0.44\sigma_0)$$

so,

$$\frac{\left(\frac{dN_A}{dt}\right)_e}{\left(\frac{dN_A}{dt}\right)_{th}} \leq \frac{2}{R-1} \quad (24)$$

If the cadmium ratio is larger or equal to 200, we find that the contribution to the total induced activity due to epithermal neutrons is less than 1% of the activity induced by thermal neutrons. In such a case the accuracy of the analysis will be practically unaffected by even the existence of a serious absorption effect due to epithermal neutrons.

It must be pointed out that the cadmium ratio given in Eq. (24) is for the nuclide which gives rise to the measured induced activity. Very often the cadmium ratio for an irradiation position is given or measured for gold. By the use of Eq. (11) we can find the cadmium ratio for any nuclide expressed in terms of the cadmium ratio for gold:

$$R = 1 + (R^{Au}-1) \frac{(J^{Au} + 0.44\sigma_0^{Au})\sigma_0}{(J + 0.44\sigma_0)\sigma_0^{Au}} \quad (25)$$

Measured and calculated values for  $(J+0.44\sigma_0)$  are tabulated in a compilation by Macklin and Pomerance.<sup>12</sup> (The data are also reproduced in "Reactor Physics Constants."<sup>4</sup>) Data for  $\sigma_0$  can be found in "Neutron Cross-Section Data."<sup>24,25</sup>

In order to determine the cadmium ratio for an irradiation position, one must either use dilute solutions or very thin foils in which no absorption effects are present, or correct all measured activities for these effects.

To illustrate the effects of neutron absorption on the determination of cadmium ratios we will mention that the uncorrected cadmium ratio for gold in one of the pneumatic tubes of the Ford Nuclear Reactor of The University of Michigan is 12.5 using 1 mil thick gold foil, while the cadmium ratio found by using a dilute solution of gold ( $7.2 \times 10^{-2}$  mg gold/ml) is 5.5. The difference is primarily due to the effect of absorption of epithermal neutrons. If gold foils are used for determining cadmium ratios, absorption corrections<sup>1</sup> are needed if the gold thickness exceeds about 0.5 mg/cm<sup>2</sup>.

A discussion of absorption effects in foils can be found in the literature.<sup>4-7,26-28</sup>

### 3.7. CALCULATIONS OF EFFECTIVE RESONANCE INTEGRALS (Determination of Copper and Gold in Silver)

It is very difficult to give general rules for calculating  $J^{\text{eff}}$  in Eq. (15) [or the integral in Eq. (22)] for a specific nuclide in a particular sample. The calculations will depend on the nuclide of interest and on the composition of the sample.

We can divide all neutron activation analysis into two groups. The first group contains cases where the main absorption resonances in the nuclide of

interest overlap partially or completely with main resonances in the nuclides which make up the main constituents of the sample. The second group represent cases where no overlapping of resonances occur.

In order to illustrate the kind of calculations involved we will in the present section calculate the absorption effects, and  $J^{\text{eff}}$  in particular, for gold and copper in a silver alloy, and compare the results with experimental data. Gold and copper in silver give an example from each of the two groups referred to above.

### 3.7.1 Experimental

Spheres, weighing from 8.0 to 1912 mg, were made from rods of a silver alloy containing about 1 weight-% of both copper and gold. The spheres, which were machined on a lathe, were etched in nitric acid, washed in distilled water, and weighed. The measured radii of the spheres were compared with the radii calculated from the weights to exclude the possibility of caves in the spheres. The homogeneity of the silver alloy with respect to copper and gold was checked and found good by the emission spectrographic division of the Central Institute for Industrial Research, Norway.

Solutions containing 0.67, 0.10, 0.067 and 0.0134 mg gold/ml were used as gold standards. Pellets of pure copper, weighing about 30 mg, were used as copper standards.

Five spheres, together with copper standards and four quartz ampoules containing the four gold solutions were packed in an aluminum can and irradiated with pile neutrons from the reactor "JEEPPI", Norway. A similar can was also irradiated in a thermal column of the same reactor. The cadmium

ratios for gold at the two irradiation positions were 2.6 and 11400, respectively. The flux gradient at the place of irradiation where the cadmium ratio for gold was 2.6 was found to be 0.8%/cm along the center line of the aluminum can. No flux gradient could be found in a plane normal to the center line. No measurements of the flux gradient at the irradiation position in the thermal column were made. The standards were placed so as to minimize the effects of inhomogeneities in the neutron flux.

It is not recommended to irradiate many heavy absorbing samples together. It might happen that one or more samples are lying in the perturbed flux caused by other absorbers, or in the "shadow" from other samples caused by a possible streaming of neutrons in the isotope channel. Even if the samples and standards in the present experiment were packed with the possibility of these effects in mind, it is not unlikely that some of the differences between the results of the copper and gold content in the different spheres can be accounted for by these effects.

Copper and gold from the silver alloy were radiochemically separated after the irradiation and the gross gamma activities were measured with a NaI(Tl) well crystal.<sup>15</sup> The gross gamma activities of the standards were measured under the same conditions without chemically separating the copper and gold. The 12.8 hour  $\text{Cu}^{64}$  activity was measured for the determination of copper.

### 3.7.2 Theory

The specific activity of the four gold solutions, which were irradiated in the thermal column, was constant. When the solutions were irradiated

with pile neutrons, the most concentrated solution (0.67 mg Au/ml) showed a specific activity which was 5% below the specific activity of the other three solutions. The result of the specific activity of this standard was not used. This decrease in the specific activity is most probably due to a resonance absorption effect of neutrons with energies around 4.91 eV.

Because there are no absorption effects in the standards, the ratio of the measured activity of copper and gold from a sphere to the measured activity of the copper and gold standards, respectively, can be found from Eqs.

(10) and (18):

$$\frac{A_t}{A_t^s} = \frac{x}{p} \cdot \frac{\sigma_{\text{pile}}^{\text{eff}}}{\sigma_{\text{pile}}} \cdot \frac{\bar{N}_{\text{th}}}{N_{\text{th}}} \quad (26)$$

where  $p$  is the known amount of copper or gold in the standard. By using Eqs. (8), (11) and (17) we can write Eq. (26) in the form

$$\frac{A_t}{A_t^s} = \frac{x}{p} \left[ 1 - \frac{1}{R} + \frac{(J^{\text{eff}} + 0.44\sigma_0)}{(J + 0.44\sigma_0)R \cdot \frac{\bar{N}_{\text{th}}}{N_{\text{th}}}} \right] \frac{\bar{N}_{\text{th}}}{N_{\text{th}}} \quad (27)$$

or

$$\frac{A_t}{A_t^s} = \frac{x}{p} \cdot L \cdot \frac{\bar{N}_{\text{th}}}{N_{\text{th}}} \quad (28)$$

where

$$L = 1 - \frac{1}{R} + \frac{(J^{\text{eff}} + 0.44\sigma_0)}{(J + 0.44\sigma_0)R \cdot \frac{\bar{N}_{\text{th}}}{N_{\text{th}}}} \quad (29)$$

A cadmium ratio for gold equal to 2.6 and 11400, respectively, corresponds to a cadmium ratio for  $\text{Cu}^{63}$  equal to 32 and  $12.2 \times 10^4$ , respectively. These values can be found by using Eq. (25) and the data given in Refs. 24, 25, 29. The cadmium ratios for both  $\text{Cu}^{63}$  and gold in the thermal column are so large that the effect of the absorption of the epithermal neutrons is negligible. Instead of Eq. (27) we can, therefore, in this case use the following equations:

$$\frac{A_t}{A_s} = \frac{x}{p} \cdot \frac{\bar{N}_{th}}{N_{th}} \quad (30)$$

### 3.7.3 Calculation of $\frac{\bar{N}_{th}}{N_{th}}$ for Silver Spheres

The ratio  $\frac{\bar{N}_{th}}{N_{th}}$  for the different spheres was determined by the method outlined in Section 3.4. The absorption mean-free-path was calculated by using Eqs. (19) and (21), and values for  $\frac{\bar{N}_{th}}{N_{th}}$  were found by the curve in Figure 1.

The inorganic division of the Central Institute for Industrial Research, Norway, determined the copper and gold content in the silver alloy by a spectrophotometric and a titrimetric method, respectively. The results were given as 1.07 weight-% copper and 1.03 weight-% gold. The results from the two different rods, from which the spheres were machined, differed by 1%. The alloy was also supposed to contain magnesium, aluminum, silicon, manganese, zinc, tin, antimony and lead. The concentrations of these elements were unknown, but the silver content was determined as 94 weight-%. Since none of these elements, nor copper, have absorption cross-sections larger than the absorption cross-section for silver, the absorption mean-free-path must be smaller than the mean-free-path in a silver alloy containing 94% silver, 1% gold and

5% of elements with zero absorption cross-sections, but larger than the absorption mean-free-path in an alloy containing 98% silver, 1% gold and 1% copper. The results of the calculations are  $0.192 \sqrt{T}$  mm and  $0.182 \sqrt{T}$  mm, respectively. The true value for the mean path is likely to be more near  $0.192 \sqrt{T}$  mm than  $0.182 \sqrt{T}$  mm, but since we cannot determine the value very accurately, we have used the average value,  $0.187 \sqrt{T}$  mm in the following calculations. The neutron temperature in the "fast column" was given as 395°K. The neutron temperature in the thermal column was not known, but it was assumed to be 10°C higher than the graphite temperature, which was 45°C. This assumption was based on the fact that the neutron temperature in the thermal column of the reactor "RI," Stockholm, Sweden, which also is a heavy water moderated, inhomogeneous reactor of nearly the same type as "JEEPI," is about 10°C over the graphite temperature. The absorption mean-free-path in the spheres, which are irradiated in the "fast column," is therefore 3.7 mm, and equal to about 3.4 mm in the spheres which are irradiated in the thermal column.

Calculated values for  $\frac{\bar{N}_{th}}{N_{th}}$  for the different silver spheres used in the experiment are given in Table VI and Table VII (Section 3.7.6).

#### 3.7.4 The Effective Resonance Integral for Copper-63 in Silver

Copper-63 has no reported neutron resonances below 580 eV.<sup>24</sup> Although a sum of 31 resonances is reported<sup>24</sup> for Ag<sup>107</sup> and Ag<sup>109</sup>, no resonances are above 442 eV. Assuming a  $\frac{1}{v}$ -law for the absorption cross-section for natural silver for neutrons above 580 eV, we find that the cross-section must be less than 0.4 barn for energies above 580 eV. Considering the neutrons with an

energy greater than 580 eV, we find that the absorption mean-free-path for these neutrons must be greater than 430 mm in the silver alloy. Even in a sphere which weighs 2000 mg, the average absorption of neutrons with energies around 580 eV will be less than 1%. We can therefore disregard the effect of absorption of neutrons in the  $\text{Ag}^{107}$  and  $\text{Ag}^{109}$  nuclides on the resonance integral for  $\text{Cu}^{63}$ . But since the copper content is as high as 1 weight-%, we have to consider the possibility of a self-absorption effect on the resonance integral.

The calculations of the effective resonance integral for  $\text{Cu}^{63}$  will be based on the work of Chernick and Vernon.<sup>8</sup> They give the following equations for a single Breit-Wigner resonance (without any corrections for Doppler effect) in the "narrow resonance (NR)" approximation and in the "narrow infinitely heavy absorber (NRIA)" approximation, respectively:

$$J_i^{\text{eff}}(\text{NR}) = J_i \left[ 1 + \frac{\beta}{\epsilon+1} - \frac{\beta\Gamma_n}{\Gamma(\epsilon+1)^2} \right]^{-\frac{1}{2}} \quad (31)$$

$$J_i^{\text{eff}}(\text{NRIA}) = J_i \left[ 1 + \frac{\beta\Gamma_\gamma}{\epsilon\Gamma} \right]^{-\frac{1}{2}} \quad (32)$$

where

$$J_i = \frac{\pi\Gamma_\gamma\sigma_t}{2E_r}, \quad \beta = \frac{\sigma_t}{\sigma_p}, \quad \text{and} \quad \epsilon = \frac{\rho}{4N_0\sigma_p} \cdot \frac{S}{M}$$

and where

$\sigma_t$  = the total cross-section at the maximum of the resonance

$\sigma_p$  = potential scattering cross-section equal to  $4\pi r^2$  where  $r$  is the radius of the nucleus

$\rho$  = specific density of the lump

$N_0$  = number of absorbing nuclides/cm<sup>3</sup>

$S$  = surface area of the lump

$M$  = mass of the lump

$E_r$  = resonance energy

$\Gamma_n$  = partial width for neutron emission

$\Gamma_\gamma$  = partial width for gamma emission

$\Gamma = \Gamma_n + \Gamma_\gamma$ .

Chernick and Vernon<sup>8</sup> recommend that 10% should be added to the calculated effective resonance integral in the case of NRIA approximation.

For spheres, we can write Eqs. (31) and (32) in the forms:

$$J_i^{\text{eff}}(\text{NR}) = J_i \left[ 1 + \frac{4N_0\sigma_t r}{3(1 + \frac{4}{3} N_0\sigma_p r)} \left( 1 - \frac{\Gamma_\gamma}{3\Gamma(1 + \frac{4}{3} N_0\sigma_p r)} \right) \right]^{-\frac{1}{2}} \quad (33)$$

$$J_i^{\text{eff}}(\text{NRIA}) = J_i \left[ 1 + \frac{4\Gamma_\gamma N_0\sigma_t r}{3\Gamma} \right]^{-\frac{1}{2}} \quad (34)$$

$(J+0.44\sigma_0)$  for  $\text{Cu}^{63}$  is equal to 3.7 barn.<sup>29</sup> Because  $\sigma_0$  is 4.5 barn,<sup>24</sup> we find that  $J$  is 1.7 barn. The first resonance in  $\text{Cu}^{63}$  has the following parameters:<sup>30</sup>  $E_r = 577$  eV,  $\sigma_t = 1550$  barn,  $\Gamma = 1.3$  eV. It is assumed that  $\Gamma_\gamma = 0.4$  eV.<sup>30</sup> Using these parameters we find that the dilute resonance integral for this resonance,  $J_1 = \frac{\pi\Gamma_\gamma\sigma_t}{2E_r}$ , is 1.7 barn. The total resonance integral for  $\text{Cu}^{63}$  can therefore be considered due to the resonance at 577 eV. The maximum energy loss in a collision between a neutron with an energy of 577 eV and a copper nucleus is much greater than the practical width,  $\Gamma(\frac{\sigma_t}{\sigma_p})^{\frac{1}{2}}$ , so the effective resonance integral should be calculated by using the NR approximation, e.g., using Eq. (33). In this equation,  $N_0$  is the number of

$\text{Cu}^{63}$  nuclides/cm<sup>3</sup> of the silver alloy and  $r$  is the radius of the actual sphere. The value for  $\sigma_p$  is found by using the formula  $4\pi r^2$  where  $r = 1.5 \cdot 10^{-13} \cdot A^{1/3}$  cm.  $A$  is the mass number. By putting the known resonance parameters and the value for  $N_0 = 6.88 \cdot 10^{20}$  nuclides/cm<sup>3</sup> into Eq. (33) we get:

$$J_1^{\text{eff}}(\text{NR}) = J_1 \left[ 1 + \frac{0.142r}{1 + 0.0004r} \left( 1 - \frac{0.1025}{1 + 0.0004r} \right) \right]^{-\frac{1}{2}}$$

where  $J_1 = 1.7$  barn and where  $r$  is in mm.

Because  $r$  is less than 3.6 mm, we can set  $1 + 0.0004r$  approximately equal to 1:

$$J_1^{\text{eff}}(\text{NR}) = J_1 [1 + 0.127r]^{-\frac{1}{2}} \quad (35)$$

The values for  $J_1^{\text{eff}}(\text{NR})$  for the five spheres irradiated in the "fast column" are given in Table II.

Table II.  $J_1^{\text{eff}}(\text{NR})$  for the 577-eV Resonance in  $\text{Cu}^{63}$  as a Function of the Radius of the Silver Spheres.

Radius $r$ in mm	0	0.55	1.00	1.50	2.50	2.45
$J_1^{\text{eff}}(\text{NR})$ in barns	1.7	1.65	1.6	1.55	1.5	1.4

As pointed out earlier (Section 3.6),  $J^{\text{eff}}$  in Eq. (29) also takes into account the absorption effect on the contribution to the induced activity due to  $\frac{1}{v}$ -absorption of the epithermal neutrons. To make this clear, we will write  $(J^{\text{eff}} + 0.44\sigma_0)$  as  $(J_1^{\text{eff}} + d \cdot 0.44\sigma_0)$ , where  $J_1^{\text{eff}}$  is given in Eq. (35) (note that the total resonance integral is very close to  $J_1$ ) and where  $d$  is a correction

factor which takes into account the absorption effect on the  $\frac{1}{v}$ -induced activity, caused by epithermal neutron absorption in silver nuclides. We have seen that the absorption of neutrons with energies above 580 eV is unimportant, so when we are calculating the factor  $d$  we have only to consider the region 0.4 eV-580 eV which contains a sum of 31 reported neutron resonances in  $\text{Ag}^{107}$  and  $\text{Ag}^{109}$ . Let us proceed by discussing first how  $d$  depends on the resonance absorption in  $\text{Ag}^{107}$  and  $\text{Ag}^{109}$ , and then the effect of the  $\frac{1}{v}$ -absorption in the silver nuclides.

The most important resonance occurs in  $\text{Ag}^{109}$  at an energy of 5.20 eV. The absorption cross-section at peak maximum is about  $3.1 \times 10^4$  barns,<sup>31</sup> and neutrons with energies close to 5.2 eV will essentially be completely absorbed in a thin layer at the surface of the spheres. The next highest resonance occurs in  $\text{Ag}^{107}$  at an energy of 16.6 eV. The absorption cross-section at peak maximum is about 2650 barns.<sup>32,33</sup> The  $\text{Cu}^{63}$  induced activity due to  $\frac{1}{v}$ -absorption of neutrons within the range 0.4-17 eV is responsible for about 95% of all the  $\frac{1}{v}$ -induced activity due to epithermal neutrons. We will therefore only look at the resonance absorption effect due to the two resonances at 5.2 and 16.6 eV. If we assume that all neutrons within the ranges 5.05-5.35 eV and 16.4-16.8 eV, which correspond to about two times the total width,  $\Gamma$ , for the two resonances, are completely removed from the flux, the  $\frac{1}{v}$ -induced activity will only decrease by 1.2% with respect to the activity induced by an unperturbed flux. We will therefore consider the factor  $d$  as independent of the resonance absorption in  $\text{Ag}^{109}$  and  $\text{Ag}^{107}$ .

To take the effect of the  $\frac{1}{v}$ -absorption of neutrons in the silver nuclides

into consideration, we will set the correction factor  $d$  equal to the ratio of the average density of neutrons, within the range 0.4-16 eV, in a silver sphere with a radius  $r$  mm to the unperturbed density of neutrons, within the same energy range ( $d = \frac{\Delta n_e}{\Delta n_e}$ ). In calculating the average cross-section for this region we will assume that the cross-section follows the  $\frac{1}{v}$ -law. Because the number of neutrons within the range 5.0-5.3 eV is small compared to the total number of epithermal neutrons, it is not necessary to consider the 5.2 eV resonance. By using Eqs. (19) and (20) we find  $\bar{\sigma} = 7.7$  barn and  $l = 24$  mm. Values for  $d = \frac{\Delta n_e}{\Delta n_e}$  for the different spheres used in the determinations are tabulated in Table III together with values for  $(J_1^{\text{eff}} + d \cdot 0.44 \sigma_0) = (J^{\text{eff}} + 0.44 \sigma_0)$ .

Table III. The Correction Factor  $d$  (Defined in the Text) and  $(J^{\text{eff}} + 0.44 \sigma_0)$  for  $\text{Cu}^{63}$  for the Different Silver Spheres Used.

Radius in mm	0	0.55	1.00	1.50	2.50	3.45
$d$	1.00	0.99	0.97	0.96	0.93	0.90
$(J^{\text{eff}} + 0.44 \sigma_0)$ barn	3.7	3.6	3.5	3.45	3.3	3.2

### 3.7.5 The Effective Resonance Integral for Gold-197 in Silver

We will now calculate  $(J^{\text{eff}} + 0.44 \sigma_0)$  for gold. This monoisotopic element has one large neutron resonance at an energy of 4.91 eV and a smaller one at 16.5 eV. These two resonances are responsible for more than 97% of the total dilute resonance integral for gold, and the other small resonances will not be considered. The resonance at 4.91 eV has the following parameters:<sup>34</sup>

$E_r = 4.906$  eV,  $\Gamma_\gamma = (0.124 \pm 0.003)$  eV,  $\Gamma_n = (0.0156 \pm 0.0004)$  eV,  $\sigma_t = (37,000 \pm 500)$  barn, and  $\sigma_p = (11.1 \pm 0.8)$  barn. The dilute resonance integral for this resonance,  $J_1$ , is equal to  $(1468 \pm 44)$  barn. The resonance at 16.5 eV has the following parameters:<sup>32</sup>  $E_r = 61.5$  eV,  $\sigma_t \Gamma^2 = (450 \pm 130)$  barn, and  $\Gamma_\gamma$  is assumed to be 0.15 eV. Hughes and Harway<sup>24</sup> give the value  $\Gamma_n = (110 \pm 20)$  mV and  $\Gamma_\gamma = (170 \pm 80)$  mV. By using the value  $\Gamma_\gamma = 0.15$  eV, we find that  $\Gamma$  is about 0.25 eV and that  $\sigma_t$  is about 6700 barn. The dilute resonance integral for this resonance,  $J_2$ , is therefore about 25 barn. The uncertainty in  $J_2$  is probably of the order of  $\pm 30\%$ .  $(J + 0.44\sigma_0)$  is reported to be 1558 barn,<sup>12</sup> and  $\sigma_0 = (98.8 \pm 0.3)$  barn,<sup>24</sup> so  $J$  is 1515 barn.  $J_1 + J_2$  is 1493 barn, and we therefore have the possibility that the rest of the resonances are responsible for about 20 barn. But the uncertainty in  $J$ ,  $J_1$  and particularly in  $J_2$  is so great that this cannot be a definite conclusion. It is also possible that the resonance at 4.9 eV is responsible for nearly 100% of the total resonance integral, or that  $J_1$  is close to the calculated value, 1468 barn, and that  $J_2$  takes care of the rest of the total resonance integral, that is  $J_2 = 47$  barn.

Let us write  $J = J_1 + J_2 + \sum J_i$ , and  $(J^{\text{eff}} + 0.44\sigma_0) = (J_1^{\text{eff}} + J_2^{\text{eff}} + \sum J_i^{\text{eff}} + d \cdot 0.44\sigma_0)$ , where  $d$  is the correction factor defined in the case of copper in silver. By using the same arguments as in the copper case we can find that the values for  $d$  will be the same as those listed in Table III. Because of the uncertainty in  $J_1$ ,  $J_2$ , and  $\sum J_i$ , we will calculate  $(J^{\text{eff}} + 0.44\sigma_0)$  for two extreme cases, A and B, where we only consider changes in the resonance integral  $J_1$ . We will write  $(J^{\text{eff}} + 0.44\sigma_0)$  equal to  $(J_1^{\text{eff}} + J_3 + d \cdot 0.44\sigma_0)$ , where in case A we will have  $J_1 = 1515$  barn and  $J_3 = 0$  barn, and in case B,  $J_1 = 1468$  barn and

$J_3 = 47$  barn. The values for  $(J_1^{\text{eff}} + 0.44\sigma_0)$  in the two cases will be labelled  $(J_1^{\text{eff}} + 0.44\sigma_0)_A$  and  $(J_1^{\text{eff}} + 0.44\sigma_0)_B$  respectively. The two values for  $(J_1^{\text{eff}} + 0.44\sigma_0)$  in the two cases will give us a maximum and a minimum value because of the fact that the greatest effect of the neutron absorption on  $(J_1^{\text{eff}} + 0.44\sigma_0)$  will be due to the change in  $J_1^{\text{eff}}$  caused by resonance absorption of neutrons with energies around 5.2 eV in  $\text{Ag}^{109}$ .

It is very difficult to calculate  $J_1^{\text{eff}}$  accurately in this case but the resonance in gold at 4.91 eV overlaps the resonance in  $\text{Ag}^{109}$  at 5.2 eV to a great extent and it is reasonable to assume that  $J_1^{\text{eff}}$  for gold will decrease with the same rate as the effective resonance integral for silver:

$$\left(\frac{J_1^{\text{eff}}}{J_1}\right)_{\text{Au}} = \left(\frac{J_1^{\text{eff}}}{J_1}\right)_{\text{Ag}}$$

The neutron resonance in  $\text{Ag}^{109}$  at 5.2 eV has the following parameters:<sup>31</sup>  
 $E_r = (5.199 \pm 0.010)$  eV,  $\sigma_t = (34,000 \pm 1000)$  barn,  $\sigma_p = (4.7 \pm 0.8)$  barn,  
 $\Gamma = (0.149 \pm 0.005)$  eV,  $\Gamma_n = (0.0134 \pm 0.006)$  eV, and  $\Gamma_\gamma = (0.136 \pm 0.006)$  eV.  
The practical width,  $\Gamma\left(\frac{\sigma_t}{\sigma_p}\right)^{\frac{1}{2}}$ , is about 12.7 eV, while the maximum energy loss in a collision between a neutron with an energy of 5.2 eV and a silver nucleus is about 0.2 eV. We can therefore calculate  $\left(\frac{J_1^{\text{eff}}}{J_1}\right)_{\text{Ag}}$  by using the NR1A approximation, e.g., using Eq. (34). By putting the appropriate resonance parameters and the value  $2.85 \times 10^{22}$  for the number of  $\text{Ag}^{109}$  nuclides/cm<sup>3</sup>,  $N_0$ , into this equation, we get:

$$\left(\frac{J_1^{\text{eff}}}{J_1}\right)_{\text{Ag}} = [1 + 118r]^{-\frac{1}{2}} \quad (36)$$

where  $r$  is in mm. Values for  $(\frac{J_1^{\text{eff}}}{J_1})_{\text{Ag}}$  for the five different spheres used in the gold determinations are given in Table IV, where 10% is added to the values calculated by Eq. (36) as recommended by Chernick and Vernon.<sup>8</sup>

Table IV.  $(\frac{J_1^{\text{eff}}}{J_1})_{\text{Ag}}$  for the Neutron Resonance at 5.199 eV as a Function of the Radius of the Silver Spheres.

Radius in mm	0	0.55	1.00	1.50	2.50	3.45
$(\frac{J_1^{\text{eff}}}{J_1})_{\text{Ag}}$	1.000	0.13	0.10	0.082	0.064	0.055

In Table V we have given the values for  $(J^{\text{eff}+0.44\sigma_0})_A$  and  $(J^{\text{eff}+0.44\sigma_0})_B$ , which represent the two extreme cases discussed above. The average of the two values is also given.

Table V.  $(J^{\text{eff}+0.44\sigma_0})$  for Gold as a Function of the Radius of the Spheres in the Two Cases, A and B, Discussed in the Text.

Radius in mm	0	0.55	1.00	1.50	2.50	3.45
$(J^{\text{eff}+0.44\sigma_0})_A$ in barns	1558	270	194	166	132	112
$(J^{\text{eff}+0.44\sigma_0})_B$ in barns	1558	281	236	209	181	167
$(J^{\text{eff}+0.44\sigma_0})_{\text{avg}}$ in barns	1558	260	215	188	156	140

We have used the average value  $(J^{\text{eff}+0.44\sigma_0})_{\text{avg}}$ , in the calculation of the correction factor,  $L$ , defined in Eq. (29). To see the effect of the difference between  $(J^{\text{eff}+0.44\sigma_0})_A$  and  $(J^{\text{eff}+0.44\sigma_0})_B$  on  $L$ , we have calculated  $L$  for the smallest and the largest sphere by using the value for  $(J^{\text{eff}+0.44\sigma_0})$

in the two cases, A and B. For the smallest sphere,  $r = 0.55$  mm, the difference between the two values of  $L$  is only 1.6%, and for the largest sphere,  $r = 3.45$  mm, the difference is still only 3.7%.

### 3.7.6 Results

I. Thermal Column. The values for the concentration of copper and gold in the silver alloy spheres, which were irradiated in the thermal column, are given in Table VI. Values of  $\frac{\bar{N}_{th}}{N_{th}}$  are also included. Equation (30) was used in the calculations. Values for  $\frac{\bar{N}_{th}}{N_{th}}$  were found as described in Section 3.4. The error given for the average value is only the standard deviation for the five different values given and includes no errors in  $\frac{\bar{N}_{th}}{N_{th}}$ .

Table VI. Concentration of Copper and Gold in Five Silver Spheres Irradiated in the Thermal Column ( $R^{Au} = 11,400$ ).

Weight (mg)	Radius in mm		$\frac{\bar{N}_{th}}{N_{th}}$	Weight-% Cu	Weight-% Au
	Measured	Calculated			
1912	3.55	3.54	0.48	1.08	1.13
704.3	2.60	2.54	0.58	0.96	0.96
155.6	1.55	1.53	0.73	0.90	0.91
48.38	1.00	1.01	0.81	1.10	0.98
10.20	0.60	0.58	0.88	1.11	1.04
Average:				$1.03 \pm 0.09$	$1.00 \pm 0.08$

II. Fast Column. The values for the concentration of copper and gold in the silver alloy spheres, which were irradiated with pile neutrons, are given in Table VII. Equations (28) and (29) were used in the calculations.

Values for  $\frac{\bar{N}_{th}}{N_{th}}$  and  $L$  are also included. The error given for the average value is only the standard deviation for the five different values given, and does not include errors in  $\frac{\bar{N}_{th}}{N_{th}}$  nor in  $L_{Cu}$  and  $L_{Au}$ .

Table VII. Concentration of Copper and Gold in Five Silver Spheres Irradiated in the "Fast Column" ( $R^{Au} = 2.6$ ).

Weight (mg)	Radius in mm		$\frac{\bar{N}_{th}}{N_{th}}$	$L_{Cu}$	$L_{Au}$	Weight-% Cu	Weight-% Au
	Measured	Calculated					
1794	3.45	3.46	0.52	1.02	0.68	1.08	1.12
703.6	2.50	2.53	0.61	1.01	0.68	1.01	0.99
149.2	1.50	1.50	0.75	1.01	0.68	0.86	0.97
48.11	1.00	1.03	0.82	1.005	0.68	0.92	0.97
8.00	0.55	0.56	0.90	1.00	0.69	1.20	0.91
Average:						1.01±0.14	0.99±0.06

III. Standard Analytical Methods. As already mentioned, the concentration of copper and gold in the silver alloy was determined by a spectrophotometric and a titrimetric method, respectively. The results were given as 1.07 weight-% Cu and 1.03 weight-% Au.

### 3.7.7 Discussion

It should be pointed out that the analysis of copper and gold in the silver alloy by the method reported were carried out only to illustrate the effect of the neutron absorption and to show how to approximately calculate these effects. In practice, the experimental part of a neutron activation analysis should, if possible, be designed so that the absorption effects will

be eliminated or experimentally determined.

It is important to notice, in Table IV or by Eq. (36), that the ratio  $\left(\frac{J_1^{\text{eff}}}{J_1}\right)_{\text{Ag}}$  decreases very fast with the sample size, for spheres with radii less than a few tenth of a mm, while the same ratio, for spheres with radii larger than about 0.5 mm, decrease much slower with the sample size. As a consequence of this, the effect of the absorption of the epithermal neutrons (actually neutrons with energies around 5 eV) on the gold determinations, is great and almost the same for the sample weighing 8.00 mg as for the sample weighing 1794 mg.

The rapid decrease in the effective resonance integral for gold in silver is caused mainly by resonance absorption in  $\text{Ag}^{109}$ , and only to a small extent by self absorption in the gold nuclides. We will calculate the effect of this self absorption by calculating the effective resonance integral for gold under the assumption that no neutrons are absorbed in the 5.2 eV resonance in  $\text{Ag}^{109}$ . We will only calculate the effective resonance integral for the first large resonance at 4.91 eV, and we will use the value 1515 barn for the dilute resonance integral for this resonance (case A above).

The effective resonance integral for this wide low-lying resonance can be treated in the NRIA approximation, and by using Eq. (34) and the resonance parameters given above, we find:

$$J_1^{\text{eff}}(\text{NRIA}) = J_1 [1 + 1.40r]^{-\frac{1}{2}} \quad (37)$$

where  $J_1 = 1515$  barn and  $r$  is in mm.

The values for  $J_1^{\text{eff}}(\text{NRIA})$  for the five different spheres used in the experiments are given in Table VIII. Ten percent is added to the calculated values for  $J_1^{\text{eff}}(\text{NRIA})$  as recommended.<sup>8</sup> In the same table we have also included corresponding values for L.

Table VIII.  $J_1^{\text{eff}}(\text{NRIA})$  for the 4.91 eV Resonance in Gold, and the Corresponding Values for L in the Case of no Interference From the 5.2 eV Resonance in  $\text{Ag}^{109}$  (1 Weight-% Gold in Silver).

Radius (mm)	0	0.55	1.00	1.50	2.50	3.45
$J_1^{\text{eff}}(\text{NRIA})$ , barns	1515	1254	1076	946	787	690
L	1.00	0.97	0.96	0.95	0.96	0.97

The table shows that the  $J_1^{\text{eff}}$  for gold, caused by a self absorption effect, decreases appreciably with the sample size, but that the effect on the correction factor, L, is small. The reason for this is that  $\frac{\bar{N}_{\text{th}}}{N_{\text{th}}}$  for the silver spheres decreases with nearly the same velocity as  $J_1^{\text{eff}}$ .

It will be important for the discussion in the next section to consider determinations of elements which have large resonance peaks, in samples which have small thermal absorption cross-sections, and which have no resonance peaks which overlap the resonance peaks in the element which are going to be determined. Let us assume that we have three iron samples, in the form of spheres, which contain 0.01, 0.1 and 1 weight-% gold, respectively. We will further assume that we use dilute gold solutions as standards such that we can use Eqs. (28) and (29) in our calculations. The cadmium ratio for gold and the neutron temperature will be assumed to be 2.6 and 400°K, respectively.

Values for  $\frac{\bar{N}_{th}}{N_{th}}$  and L for samples weighing 100, 500, and 1000 mg, are given in Table IX.

If we want to determine gold in iron (steel) samples and we want to spike some of the iron samples with gold as gold standards, we can find from Table IX that the concentration of gold must be in the range 0.05-0.01 weight-%, if the irradiations are carried out at a place where the cadmium ratio for gold is as low as 2.6.

It is interesting at this point to remember that the gold solutions used as standards in the determination of gold in silver all showed a constant specific gold activity, except for the solution containing 0.67 mg gold/ml, which had a specific activity 5% too low. This concentration corresponds to 0.067 weight-% gold in water.

#### 4. EXPERIMENTAL METHODS FOR ELIMINATION OF ABSORPTION EFFECTS

As pointed out before, the problem of calculating the effects of neutron absorption in a pile neutron activation analysis is often so complex so as to prohibit an accurate theoretical treatment. If possible, the analysis should be designed so that the problems of neutron absorption are avoided.

It is too difficult to write down a compilation of methods by which the experimentalist in a special case can choose one which will give results corrected for all absorption effects. The method used will depend on the chemical composition of the sample, its physical state (lumps, powder, solutions, etc.), the irradiation facilities (a fixed position in a pneumatic tube, or a variable position in an isotope channel), and the degree of accuracy wanted.

Table IX. Values for  $\bar{N}_{th}$  and L for Three Different Concentrations of Gold in Spheres of Iron.

Weight (mg)	Radius (mm)	$\frac{\bar{N}_{th}}{N_{th}}$ l=56 mm	$\mu\text{gm aunts. of Au}$		0.01% Au		0.10% Au		1.00% Au	
			$\frac{\mu\text{gm aunts. of Au}}{j_{eff}+0.44\sigma_0}$	L	$\frac{\mu\text{gm aunts. of Au}}{j_{eff}+0.44\sigma_0}$	L	$\frac{\mu\text{gm aunts. of Au}}{j_{eff}+0.44\sigma_0}$	L	$\frac{\mu\text{gm aunts. of Au}}{j_{eff}+0.44\sigma_0}$	L
1000	3.12	0.96	1558	1.015	1534	1.01	1358	0.96	776	0.81
500	2.48	0.97	1558	1.01	1438	1.005	1393	0.97	841	0.83
100	1.45	0.98	1558	1.01	1548	1.005	1455	0.98	1001	0.87
0	0	1.00	1558	1.00	1558	1.00	1558	1.00	1558	1.00

In the following we will describe several of the methods described in the literature and point out their limitations.

#### 4.1 METHOD OF DILUTION

Effects of neutron absorption can be eliminated by using small samples and standards or diluting them by materials which have low absorption cross-sections for pile neutrons (water, graphite, sugar, etc.). Cabell and Smales<sup>35</sup> observed absorption effects in powder of cesium chloride, and Atkins and Smales<sup>36</sup> observed the same effect in powder of tantalum and tungsten oxide. By using dilute solutions of the different compounds, no effects could be observed. The absorption cross-sections for these elements are so large that one has to expect that the ratio  $\frac{\bar{N}_{th}}{N_{th}}$  will be significantly smaller than unity even for samples weighing about 10 mg. We do not have to specify the cadmium ratio at the place of irradiation because the value for  $\frac{\bar{N}_{th}}{N_{th}}$  changes slowly with the cadmium ratio.

If we want to decide when it is necessary to dilute the samples or standards in order to eliminate absorption effect, we must not only consider the effect of the absorption of the thermal neutrons, but also consider the effect of the absorption of epithermal neutrons. A consideration of antimony as a standard will emphasize this point. A calculation of  $\frac{\bar{N}_{th}}{N_{th}}$  shows that we can use up to about 200 mg of antimony as a sphere without having more than 2% absorption of thermal neutrons. But both  $Sb^{121}$  and  $Sb^{123}$  have large resonance peaks in the lower part of the epithermal region (5-21 eV) and one must expect to have effects of the absorption of these resonance neutrons if the targets are irradiated at a place where the cadmium ratio for gold is less

than about 20. This effect has been experimentally verified by Plumb and Lewis<sup>37</sup> and Høgdahl.<sup>15</sup> Høgdahl irradiated four spheres of antimony and two standard solutions of antimony containing 0.10 mg and 0.50 mg antimony/ml, respectively. The samples and standards were irradiated at a place where the cadmium ratio for gold was 2.6. From Eq. (25) and the values given for the resonance integral of  $\text{Sb}^{121}$  and  $\text{Sb}^{123}$  in Ref. 12, we find that this cadmium ratio for gold corresponds to a cadmium ratio for  $\text{Sb}^{121}$  and  $\text{Sb}^{123}$  equal to 2.1 and  $\sim 1.5$  respectively. The specific activities of  $\text{Sb}^{122}$  and  $\text{Sb}^{124}$  in the standards were constant with respect to the two standard solutions. The specific activities of  $\text{Sb}^{122}$  and  $\text{Sb}^{124}$  in the spheres, which weighed from 8.3 mg to 281.6 mg, were from 65% to 45% lower than the corresponding specific activities of the standards. The activities were measured by dissolving the spheres, evaporating aliquots of the dilute solutions of the spheres and standards on thin aluminum foils and using a GM-tube. The decay curves were resolved in the two activities of  $\text{Sb}^{122}$  and  $\text{Sb}^{124}$ . The results of the experiment are shown in Table X. The values given for  $\frac{\bar{N}_{th}}{N_{th}}$  are found from Figure 1. The values for  $K (\text{Sb}^{122})$  are the ratio between the specific activity of  $\text{Sb}^{122}$  in the spheres to the specific activity of  $\text{Sb}^{122}$  in the standard.  $K (\text{Sb}^{124})$  is the same ratio for the activity of  $\text{Sb}^{124}$ . Because  $\frac{\bar{N}_{th}}{N_{th}}$  is essential unity, the values for  $K$  are approximately equal to the correction factor,  $L$ , defined in Eq. (29).

Table X.  $\frac{\bar{N}_{th}}{N_{th}}$ ,  $K$  ( $Sb^{122}$ ), and  $K$  ( $Sb^{124}$ ) for Four Spheres of Antimony Irradiated at a Cadmium Ratio of Gold Equal to 2.6.

Weight (mg)	Radius (mm)	$\frac{r}{l}$ ( $l=73$ mm)	$\frac{\bar{N}_{th}}{N_{th}}$	$K$ ( $Sb^{122}$ )	$K$ ( $Sb^{124}$ )
281.6	2.15	0.03	0.98	0.56	0.55
96.70	1.50	0.02	0.99	0.59	0.59
65.89	1.30	0.02	0.99	0.59	0.59
8.30	0.65	0.01	0.99	0.66	0.64
0	0	0	1.00	1.00	1.00

If, therefore, we use a thermal column where the cadmium ratio is large ( $> 100$ ), we can use about 100 mg of antimony as a standard without introducing any errors due to absorption effects. By the cadmium ratio we mean either the cadmium ratio for  $Sb^{121}$  or  $Sb^{123}$ , depending on which activity is measured. But if we irradiate at a place where the cadmium ratio is so low that the epithermal neutrons contribute appreciably to the total induced activity, we must use dilute solution of antimony in order to eliminate the effects of absorption of resonance neutrons in  $Sb^{121}$  and  $Sb^{123}$ .

Plumb and Lewis<sup>37</sup> irradiated spheres of antimony in two irradiation positions in ORNL-X-10, Oak Ridge. They give no values for the cadmium ratios. One of the positions is referred to as a "relatively cool low-flux region," and it is possible that they are primarily measuring the small effect of the absorption of the thermal neutrons in this position. For the other position they show the great effect of the absorption of resonance neutrons in  $Sb^{121}$ . From the data given in their paper,<sup>37</sup> it has been impossible for the author

to prove their statement that the attenuation effect in  $\text{Sb}^{122}$  is much larger than that for  $\text{Sb}^{123}$ . Their statement is in contradiction to the results given in Table X, from which we find that the absorption effects are the same for the two isotopes.

#### 4.2 INTERNAL STANDARD: METHOD I

It is possible to make up a standard such that the effects of neutron absorption in the standards and the samples are approximately the same. This can be done by adding a known amount of the element of interest to a matrix which is equal to the sample when chemical composition, weight, and geometrical form are concerned. This method, which can only be used if homogeneous mixing of the element with the sample is feasible, has been used by several investigators. As an example we will here refer to a work of Killick and Morris,<sup>38</sup> who determined traces of palladium in platinum. To avoid "neutron self shielding differences" between samples and standards, they mixed small known quantities of palladium with analytical samples.

The induced activity of the element under consideration in the samples and the standards can be expressed in the form of Eq. (18). If the average neutron density,  $\bar{N}_{th}$ , and the effective pile cross-section,  $\sigma_{pile}^{eff}$ , are the same in the standards and the samples, the unknown quantity can be found by using the simple Eq. (30), where  $\bar{N}_{th} = N_{th}$ .

The condition that the average neutron density must be the same in the samples and the standards not only demands that the degree of absorption of thermal neutrons must be the same, but also that the samples and the standards are exposed to the same external neutron flux. One must therefore avoid any

shadowing effect, which can occur if the samples and the standards are irradiated together in an inhomogeneous neutron flux. This can be easily understood by considering the extreme case where we have a beam of neutrons, and where we place the targets one after another in the beam. The difficulty with an inhomogeneous flux can be overcome by irradiating the targets one after another and using a small flux monitor attached to a fixed position in the irradiation can or "rabbit."

From Eqs. (11) and (17) we can see that the condition that  $\sigma_{pile}^{eff}$  must be the same in the samples and the standards only is fulfilled if the cadmium ratio and the effective resonance integral for the isotope used, and the average neutron density are the same for the standards and the samples. We have already discussed the possibilities of having the same value for  $\bar{N}_{th}$  in the targets, and we will proceed by discussing the conditions connected to the cadmium ratio and the effective resonance integral.

If the cadmium ratio changes rapidly with the space coordinates, the same cadmium ratio can be obtained by irradiating the targets one at a time and using flux monitors as described above.

Though the absorption of thermal neutrons in a sample will not be changed appreciably by adding a small quantity of the element of interest, it might well happen that the effective resonance integral will be changed. As an example we will consider the hypothetical example of determining gold in iron samples, which was discussed in Section 3.7.7 of this report. The results of the discussion were given in Table IX, and we saw that if the irradiations are carried out at a place where the cadmium ratio for gold is as low as 2.6, the

total concentration of gold in the spiked iron samples must be in the range 0.05-0.01 weight-%.

Note that the effective absorption cross-sections are approximately equal to  $\sigma_0$  if the cadmium ratio for the isotope used in the analysis is large ( $> 50-100$ ). In such cases, the only requirement is that the average neutron density must be the same in the standards and the samples.

#### 4.3 INTERNAL STANDARD: METHOD II

A similar but more sophisticated method is proposed and used by Leliaert, Hoste and Eeckhaut.<sup>39,40</sup> As in the method described in Section 4.2, they add a small known amount of the element under investigation to an analytical sample, but they use an additional element in the samples, the concentration of which can be unknown, as an internal standard. In the following we will first give the equations and statement given in their papers,<sup>39,40</sup> and then discuss under which conditions the proposed method can be used.

The element under investigation ( $c$  gm) is mixed homogeneously with one of two approximately equal samples, weighing  $a$  and  $b$  gm respectively. The two samples are irradiated together in a suitable neutron flux, and the activities induced in the internal standard and in the element of interest are measured at a time  $t$  after the end of irradiation.

The ratio of the specific activity of a suitable isotope of the standard element in the two samples is set equal to an activation correction factor,  $\alpha$ :

$$\frac{A_t(st)}{A_t'(st)} = \frac{a}{b} \cdot \alpha \quad (38)$$

where  $A_t(st)$  and  $A_t'(st)$  are the measured activities, at a time  $t$ , in the sample weighing  $a$  and  $b$  gm respectively. They now state that the same value for the correction factor,  $\alpha$ , can be applied to the element under investigation:

$$\frac{A_t}{A_t'} = \frac{ax + c}{bx} \cdot \alpha \quad (39)$$

where  $A_t$  and  $A_t'$  are the measured activities of a suitable isotope of the element under investigation at a time  $t$ , in the sample containing  $ax + c$  and  $bx$  gm of the element respectively.  $x$  is the concentration of the element in weight-%.

The above statement is true, and the method is generally useful in all cases where one can mix the added element homogeneously with the sample, provided that the cadmium ratio for the isotope of the element of interest, and which is used in the determination, is large ( $> 50-100$ ). If this is not true, some specific requirements must be added.

By using equations of the same form as Eq. (18), we get for the two ratios of the induced activities:

$$\frac{A_t(st)}{A_t'(st)} = \frac{a}{b} \left( \frac{\sigma_1^{eff}}{\sigma_2^{eff}} \right)_{st} \cdot \frac{\bar{N}_{th}}{\bar{N}_{th}'} \quad (40)$$

and

$$\frac{A_t}{A_t'} = \frac{ax + c}{bx} \cdot \frac{\sigma_1^{eff}}{\sigma_2^{eff}} \cdot \frac{\bar{N}_{th}}{\bar{N}_{th}'} \quad (41)$$

Therefore, the condition which must be fulfilled is:

$$\left( \frac{\sigma_1^{eff}}{\sigma_2^{eff}} \right)_{st} = \frac{\sigma_1^{eff}}{\sigma_2^{eff}} \quad (42)$$

By using equations of the same form as Eqs. (11) and (17), we can find that the condition in Eq. (42) is equivalent to the following requirements.

1. The cadmium ratio for the two irradiation positions occupied by the two samples must be the same.
2. The absorption effect of the thermal neutrons must be the same for the two samples:

$$\frac{\bar{N}_{th}}{N_{th}} = \frac{\bar{N}'_{th}}{N'_{th}}$$

where  $\bar{N}_{th}$  and  $\bar{N}'_{th}$  are the average thermal densities in the two samples, and where  $N_{th}$  and  $N'_{th}$  are the corresponding unperturbed neutron densities at the site of the two samples.

3. The effective resonance integral for the isotope of the element under investigation, and which is used in the determination, must be the same in the two samples. The same requirement must also apply for the effective resonance integral for the isotope used as an internal standard:

$$(J^{eff} + 0.44\sigma_0)_1 = (J^{eff} + 0.44\sigma_0)_2 \quad (43)$$

$$(J^{eff} + 0.44\sigma_0)_1^{st} = (J^{eff} + 0.44\sigma_0)_2^{st} \quad (44)$$

Conditions 1 and 2, which are normally fulfilled, require that the cadmium ratio does not change rapidly with the space coordinates, and that the two samples are approximately equal when weight and geometrical shape are concerned.

Condition 3 is fulfilled if the isotope of the element of interest, which induced activity is used in the determination, has no large resonance peaks.

If this nuclide has peaks which overlap resonance peaks in the nuclide used as an internal standard or overlap resonance peaks in the nuclides which make up the main constituents of the sample, restriction must be set on the amount of the element added. The reader is referred to the hypothetical example of determining gold in iron samples, which has been discussed in Section 3.7.7. We saw in this example that if the irradiation is carried out at a place where the cadmium ratio for gold was 2.6, the total concentration of gold in the spiked sample must be in the range 0.05-0.01 weight-%.

Though no restrictions other than the requirement that a second element which gives rise to a radioactivity measurable with good accuracy must be present in the sample are placed on the use of the method in the two reference papers,<sup>39,40</sup> Hoste, Boute, and Adams,<sup>41</sup> in a review of the method, mention the possibility that errors might be introduced if the element to be determined has large resonance peaks in the epithermal region.

Note that the activation correction factor  $\alpha$  in Eqs. (38) and (39) correct for both inhomogeneities of the neutron flux and a different absorption of thermal neutrons in the two samples.<sup>39,40</sup> Note also that it is not necessary to irradiate the two samples together but that one also can irradiate them one after another, in which case flux monitors are not needed.<sup>41</sup>

A general drawback of the methods discussed so far is that any type of manipulation of the samples prior to the activation introduce the possibility of contaminating the samples, and so invalidates one of the main advantages of activation analysis for trace constituents—namely, the elimination of reagent blanks.

#### 4.4 METHOD OF EXTRAPOLATION

Smales and Loveridge<sup>42</sup> in their determination of sodium in metallic lithium, used a general method which does not require a manipulation of the samples prior to the irradiation. They irradiated samples of different weight but with approximately the same geometrical shape, calculated the amount of sodium present without correcting for any absorption effects, and plotted the sodium content as a function of the sample size. By extrapolating to zero weight, they found the value for the sodium content corrected for absorption effect. The method requires a homogeneous neutron flux over the samples and standards, but they do point out that small variations in the flux will be smoothed out by the extrapolation process.

The method must be used with precaution. Its limitation can be illustrated by applying the method on the analysis of gold in silver, an experiment which was described in Section 3.7.

We will first apply the method on the results found by irradiating the samples in the thermal column where the cadmium ratio for gold was 11400. In this case we have only the effect of the absorption of the thermal neutrons. We have plotted in Figure 2 the values for the concentration of gold found by multiplying the values in the last column of Table VI by the value for  $\frac{\bar{N}_{th}}{N_{th}}$  as a function of the sample size. We have omitted the value for the heaviest sphere because it does not serve any important purpose in this connection. We can see from the figure that even if we have a value for the concentration of gold in the sample weighing only about 10 mg, it is very easy to introduce an error of the order of 5% by the extrapolation process. If we had only

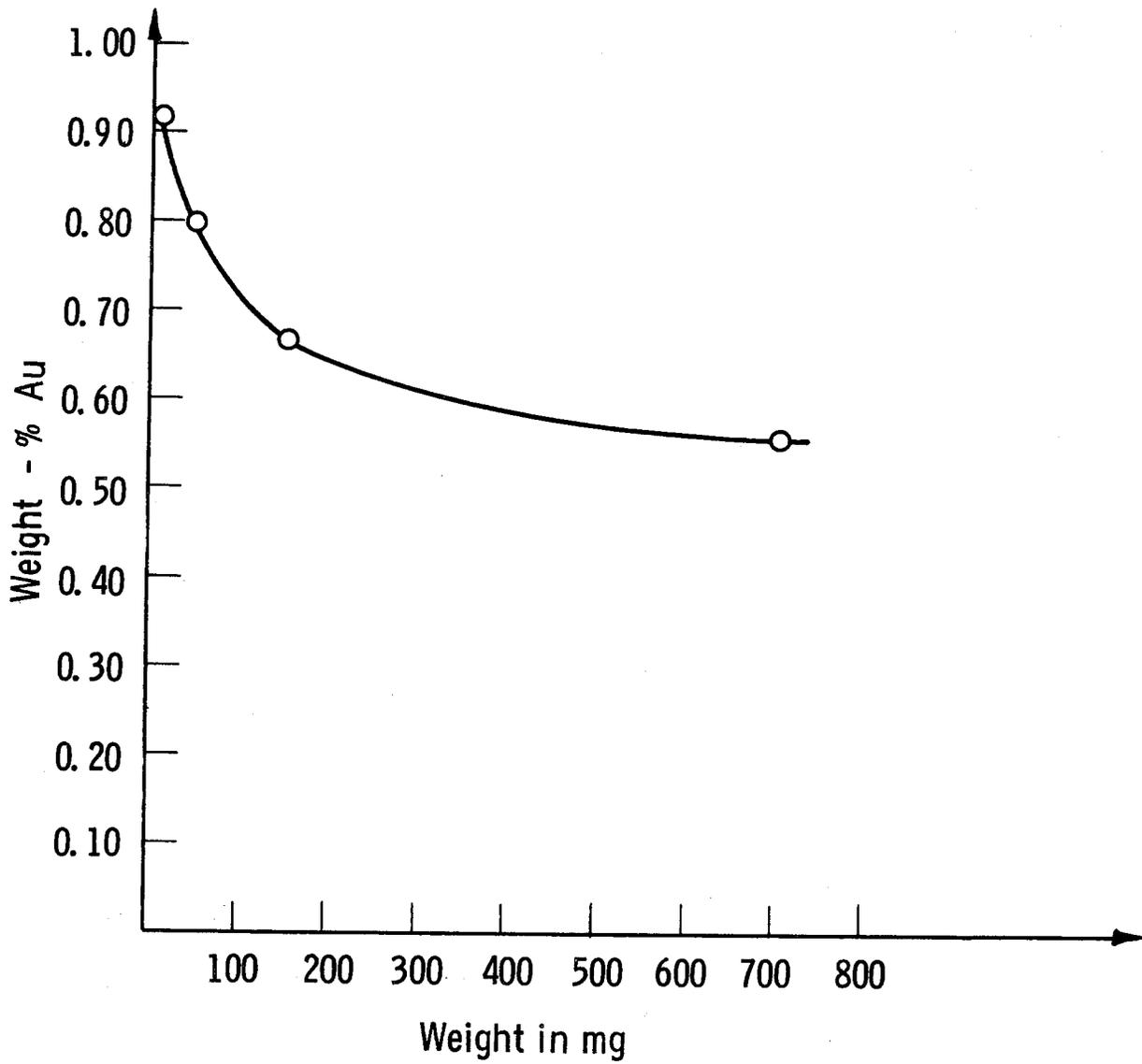


Figure 2. Concentration of gold in silver spheres found by not correcting for absorption effects ( $R^{Au} = 11400$ ). The data can be found by multiplying the values in the last column in Table VI by the values for  $\frac{N_{th}}{N_{th}}$ .

analyzed samples weighing down to about 150 mg, we could have easily introduced errors in the order of 20%.

Therefore, if the bulk part of the sample contains elements which have large thermal absorption cross-sections, it might be necessary to analyze samples weighing down to about 10 mg in order to get data good enough for an accurate extrapolation. By calculating approximately the values for  $\frac{\bar{N}_{th}}{N_{th}}$  in a given sample by the method outlined in Section 3.4, one can find approximately the minimum sample weight which is necessary for a correct extrapolation. It must be pointed out that if the sample is inhomogeneous with respect to the element under investigation, it can be very difficult and time consuming to get good values for the concentration of the element in small samples ( $\sim 10$  mg).

We will now apply the same extrapolation method on the results found by irradiating the spheres in the "fast column" ( $R^{Au} = 2.6$ ). In Figure 3, we have plotted the values for the concentration of gold found by multiplying the values in the last column in Table VII by the product of  $\frac{\bar{N}_{th}}{N_{th}}$  and  $L_{Au}$ . The plotted values are those we would have found if no corrections had been applied to the results. As in Figure 2, we have omitted the values for the heaviest sphere. It should be clear from Figure 3 that this method of extrapolation is quite inapplicable in this case. The reason for this is that the effective resonance integral for gold in samples weighing less than 10 mg decreases very rapidly with the weight of the samples. It is impossible to analyze samples so small that an accurate extrapolation can be performed. Experimentalists who are not aware of the effect of the absorption of resonance neutrons can in such cases as gold in silver, introduce an error of the order of 40% by this method.

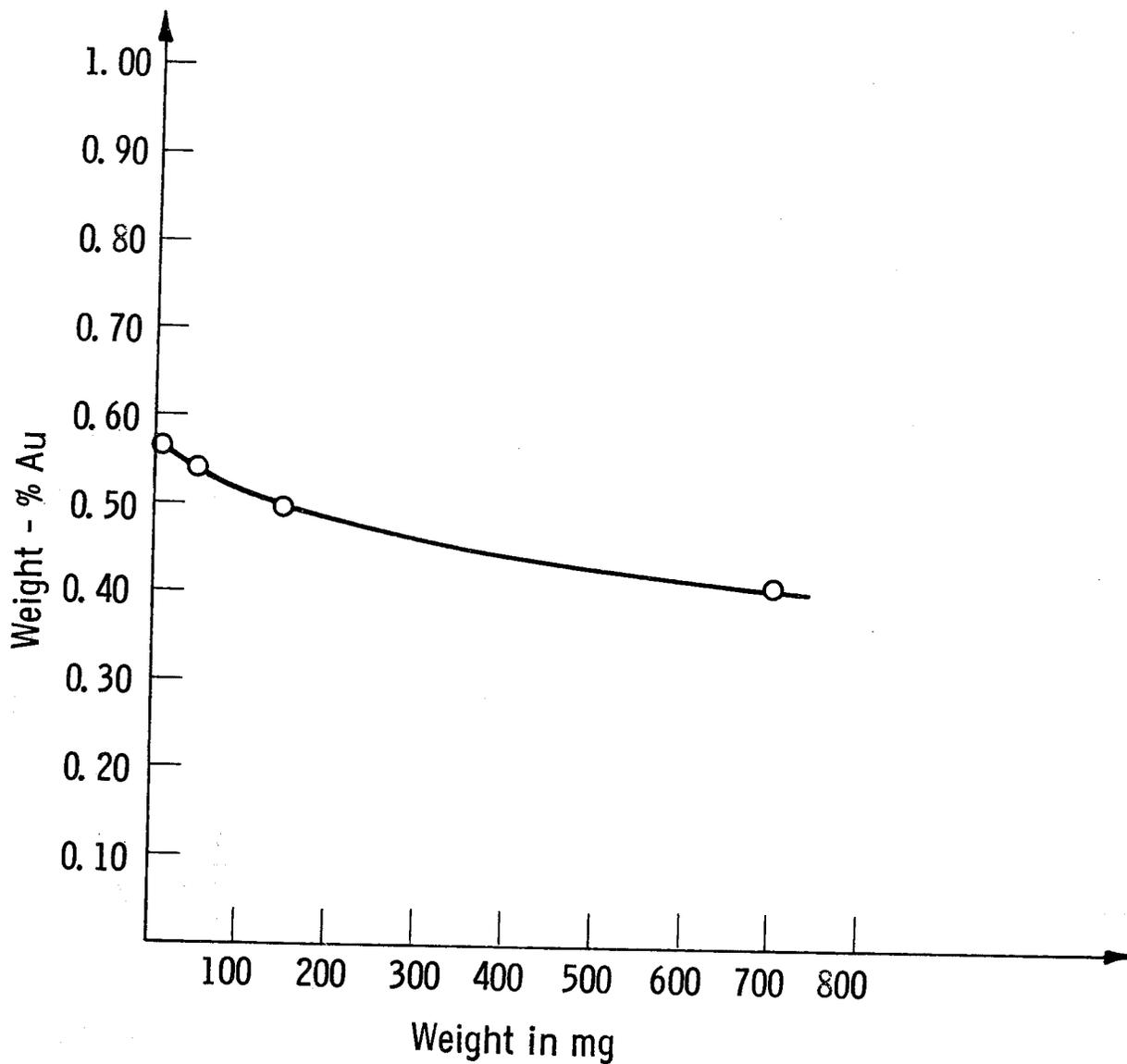


Figure 3. Concentration of gold in silver spheres found by not correcting for absorption effects ( $R^{Au} = 2.6$ ). The data can be found by multiplying the values in the last column in Table VII by the product of  $\frac{N_{th}}{N_{th}}$  and  $L_{Au}$ .

Therefore, if the bulk part of the matrix has large resonance absorption peaks which overlap those in the isotope under investigation, the method can only be used if the irradiations are carried out at a place in the reactor where the cadmium ratio is so large that the contribution to the total induced activity due to epithermal neutrons is small ( $R > 50-100$ ). Though we can get rid of the effect of the epithermal neutrons by irradiating at a place where the cadmium ratio is high, the effect of the thermal neutrons can still make it difficult to use this method.

As mentioned in the introduction to Section 4, it is very difficult to divide all examples of activation analysis into some definite groups and adopt a specific method to each of them by which the effects of neutron absorption can be eliminated. For a given analysis the experimentalist must first decide whether or not it is necessary to correct for any absorption effects, and then, if necessary, find the best method. The method which is chosen can be one of the methods already described in the literature, a combination of these, or a new method.

## 5. SUMMARY

A discussion of the absorption effects of pile neutrons in activation analysis is given. The report does not intend to give a rigorous treatment of all aspects of neutron absorption, but intends to provide the reader with a basic understanding of the absorption effects, and to give him some methods by which he can approximately calculate the absorptions effects in a given analysis.

As a basis for this discussion, the report first derives an expression for the pile neutron induced activity in a nuclide by an  $(n,\gamma)$ -reaction under the assumption of no absorption effects. The derivation is based on the assumption that the neutron spectrum is composed of a Maxwellian distribution, cut off at a velocity  $v_2$ , and an epithermal  $\frac{dE}{E}$ -distribution cut off at the same velocity. The velocity at cut off depends on the neutron temperature and on the type of reactor, but it is supposed to be less than the velocity corresponding to a neutron energy of 0.4 eV. For convenience, we have not followed the convention proposed by Westcott, Walker and Alexander,<sup>1</sup> but used a more classical convention. We have considered the induced activity as a sum of two terms, one associated with the thermal part of the spectrum (neutrons with energies up to 0.4 eV), and one term associated with the epithermal spectrum above 0.4 eV. The induced activity is expressed in terms of the density of the thermal neutrons (neutrons with energies up to 0.4 eV),  $N_{th}$ , and a pile neutron absorption cross-section,  $\sigma_{pile}$ , or a resonance integral,  $J$ . Only nuclides which follows approximately a  $\frac{1}{v}$ -law up to an energy of 0.4 eV are considered.

The effects of the absorption of pile neutrons on the neutron spectrum inside the sample and on the neutron induced activity are discussed by considering the absorption effect of the thermal neutrons ( $< 0.4$  eV) and the epithermal neutrons separately. The induced activity in the absorbing sample is expressed in terms of an average thermal neutron density,  $\bar{N}_{th}$ , and an effective pile neutron absorption cross-section,  $\sigma_{pile}^{eff}$ .

The absorption effect of the thermal neutrons is measured by the ratio

of the average thermal neutron density inside the sample to the unperturbed thermal density at the site of the sample prior to its insertion into the neutron flux,  $\frac{\bar{N}_{th}}{N_{th}}$ . The calculation of this ratio for simple geometrical forms is based on the work of Case et al.,<sup>2</sup> where the absorption mean-free-path of the neutrons is calculated by taking the assumed neutron flux distribution into consideration. Experimentally determined values of  $\frac{\bar{N}_{th}}{N_{th}}$  for spherical samples of silver, gold, cobalt and tungsten are shown to fit well with the corresponding theoretical values.

The absorption effect of the epithermal neutrons is shown to depend on the cadmium ratio at the place of irradiation and in a complex manner on the composition of the sample. The contribution to the total induced activity due to epithermal neutrons is less than 1% of the activity induced by thermal neutrons if the cadmium ratio for the nuclide of interest is larger than 200. In such a case the accuracy of the analysis will be practically unaffected by even the existence of a large absorption effect due to the epithermal neutrons.

Because of the complexity of the problem of calculating the effect due to absorption of epithermal neutrons, an accurate theoretical treatment is prohibitive. No specific rules can be given for calculating the effect because they will change from sample to sample. The calculation of the absorption effect in the activation analysis of copper and gold in silver is given as an illustration of the kind of calculations involved.

Spheres of a silver alloy containing copper and gold were irradiated with both pile neutrons and with thermal neutrons (in a thermal column). The results of the analysis were corrected for absorption effects by calculating

$\frac{\bar{N}_{th}}{N_{th}}$  and the effective resonance integral for copper and gold in silver. The average value found for five spheres irradiated in a thermal column were  $(1.03 \pm 0.09)$  weight-% copper and  $(1.00 \pm 0.08)$  weight-% gold. The corresponding values found by irradiating five spheres with pile neutrons were  $(1.01 \pm 0.14)$  weight-% copper and  $(0.99 \pm 0.06)$  weight-% gold. The errors given are only the standard deviation for the five individual values found, and do not include errors in  $\frac{\bar{N}_{th}}{N_{th}}$  or  $J^{eff}$ . The correction factors used for the analysis of the spheres irradiated in the thermal column,  $\frac{\bar{N}_{th}}{N_{th}}$ , ranged from 0.88 for the sphere weighing 10.2 mg to 0.48 for the sphere weighing 1912 mg. The correction factors used to correct the results of the copper analysis for spheres irradiated with pile neutrons (cadmium ratio for gold was 2.6) ranged from 0.90 for the sphere weighing 8.0 mg for 0.53 for a sphere weighing 1774 mg. The corresponding values for the gold analysis were 0.62 and 0.35. The concentration of copper and gold in the silver alloy was determined by a spectrophotometric and a tritrimetric method respectively. The results were given as 1.07 weight-% copper and 1.03 weight-% gold.

The analysis of copper and gold in silver by the method in this report were carried out only to illustrate the effects of the neutron absorption and to show how to approximately calculate these effects. Even if the results of the activation analysis showed a very good agreement with results found by a different analytical method, one should, if possible, design the experimental part of an activation analysis so that the absorption effects will be eliminated or experimentally determined.

Several methods by which the problem of neutron absorption can be avoided

have been used and reported in the literature. These methods have been reviewed and their limitations have been discussed.

The first method reviewed has, in this report, been called the method of dilution and consist of diluting the samples (or standards) with materials which have low absorption cross-sections for pile neutrons. In order to decide if it is necessary to dilute a sample or a standard, attention must be paid to the effects of both the thermal and epithermal neutrons. This fact was illustrated by irradiating four spheres of antimony at a place where the cadmium ratio for gold was 2.6. The experiment showed that for a sphere weighing only 8.3 mg, the specific activity of  $\text{Sb}^{122}$  and  $\text{Sb}^{124}$  were a factor of 0.66 and 0.64 respectively lower than the corresponding specific activities found by irradiating dilute solutions of antimony (0.10 and 0.50 mg antimony/ml). By calculating the ratio  $\frac{\bar{N}_{th}}{N_{th}}$  it was shown that about 100 mg of antimony can be used without introducing any absorbing effects (< 2% absorption) if the irradiations are carried out in a thermal column (cadmium ratio for gold larger than 200-300).

The second and third methods reviewed are called the internal standard I and II, respectively. In both methods a "standard" is constructed by mixing a known amount of the element of interest with one of two identical samples. But method II, which has been invented by Leliaert, Hoste and Eeckhaut,<sup>39,40</sup> make use of a second element, which concentration can be unknown, for monitoring the neutron flux in each of the two samples. The limitations of the methods are discussed in terms of the cadmium ratio and the amount of element added. A calculation shows that if the cadmium ratio for gold is 2.6,

and one wants to determine gold in a 500 mg steel sample, the amount of gold added must be less than 0.1 weight-% in order to get an error due to absorption effects less than 3%.

The fourth, and last, method reviewed is called the extrapolation method. In this method, the results of the analysis found by irradiating samples and dilute solutions of the element of interest, are plotted as a function of the sample weight. An extrapolation to zero weight gives the result corrected for absorption effects. The limitation of this method is illustrated by applying this method to the determinations of gold in silver, mentioned above. In the case where the silver spheres were irradiated in the thermal column, one must analyze samples weighing down to 10 mg in order to get the correct extrapolated value within  $\pm 5\%$ . The method is not applicable for the silver samples which were irradiated at a place where the cadmium ratio for gold was 2.6. Analysts who are not aware of the great effect of the absorption of epithermal neutrons can in this case easily introduce an error of the order of 40%.

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